

Vertical Quantum Dot Electrochemical Transistors and Complementary Inverters

Hyunwoo Jo, Seongmin Park, Hwichan Cho, Hyeono Yee, SeungHwan Roh, Myeongjae Lee, Hajin Bhang, Da In Kim, Tae Hyun Kwon, BongSoo Kim, Sohee Jeong,* and Moon Sung Kang*

Quantum dot (QD) films possess intrinsic free volumes that can be charged volumetrically with electrolytes, enabling modulation of charge density via electrochemical doping—a fundamental mechanism of electrochemical transistors (ECTs). In this work, it is reported the first demonstration of vertical QD electrochemical transistors (vQECTs), in which an *n*-type InAs QD channel is stacked vertically between the source/drain electrodes. This architecture significantly reduces the channel length to the tens of nanometer scale, offering a promising strategy to enhance device transconductance. The resulting *n*-type vQECTs exhibit a high transconductance of 20.96 (± 2.16) mS and a high integration area normalized on-current of 79.5 (± 3.54) A cm⁻² along with excellent operational stability (including endurance against bias stress, storage, and repeated on/off cycling operation). These results demonstrate performance comparable to typical *p*-type vertical organic ECTs (vOECTs), suggesting that vQECTs can serve as a complementary counterpart to existing vOECTs. To illustrate this, *n*-type vQECTs are further integrated with *p*-type vOECTs to construct vertically stacked complementary inverters, achieving a signal gain of ≈ 4.7 .

Consequently, ECTs can accumulate a higher volumetric density of charge carriers within their entire channel and attain larger signal amplification, defined via transconductance ($g_m = \partial I_D / \partial V_G$, where I_D and V_G are the drain current and gate voltage, respectively) compared to EDLTs.^[5] Based on typical transistor equations, the transconductance (most important metric) of ECTs can be defined as $g_m = (W \times d/L) \times \mu \times C^* \times (V_G - V_{th})$ in the saturation regime, where W , d , and L are the width, depth, and length of the channel, respectively, μ is the mobility, C^* is the volumetric capacitance, and V_{th} is the threshold voltage.^[1–3] Noteworthy, we refer to C^* as the volumetric capacitance because charging occurs over the entire channel volume rather than at a 2D interface (as in EDLTs). This equation suggests that g_m can be improved not only through materials engineering (improving μ or C^*)^[6–15] but also by simply controlling

the geometry of the channel (increasing $W \times d/L$).^[16–20] Although increasing d can simply increase g_m , it also degrades the switching characteristics of the device because thick channels prolong the time required for ions to penetrate or exit the entire channel.^[16,21] An alternative strategy is to shorten L . This strategy has led to growing interest in unconventional vertically structured ECTs, also known as vertical ECTs (vECTs), where the source/drain electrodes and the channel are stacked vertically.^[22–33] In vECTs, the channel length corresponds to the

1. Introduction

Electrochemical transistors (ECTs) operate through volumetric charging enabled by ions penetrating the entire body of semiconductor films under an applied electrical bias.^[1–3] This operation principle differs from that of the other-type of electrolyte-based transistors, e.g., electric double-layer transistors (EDLTs), which operate through interfacial charging induced by the formation of an EDL at the 2D semiconductor/electrolyte interface.^[4]

H. Jo, H. Cho, H. Yee, S. Roh, H. Bhang, D. I. Kim, T. H. Kwon, M. S. Kang
Department of Chemical and Biomolecular Engineering
Sogang University
Seoul 04107, Republic of Korea
E-mail: kangms@sogang.ac.kr

M. S. Kang
Institute of Emergent Materials
Sogang University
Seoul 04107, Republic of Korea

S. Park, S. Jeong
Department of Energy Science (DOES) and Center for Artificial Atoms
Sungkyunkwan University
Suwon 16419, Republic of Korea
E-mail: s.jeong@skku.edu

S. Jeong
Sungkyunkwan Institute of Energy Science and Technology (SIEST) and
Department of Display Engineering
Sungkyunkwan University
Suwon 16419, Republic of Korea

M. Lee, B. Kim
Department of Chemistry
Graduate School of Semiconductor Materials and Device Engineering
and Graduate School of Carbon Neutrality
Ulsan National Institute of Science and Technology (UNIST)
Ulsan 44919, Republic of Korea

The ORCID identification number(s) for the author(s) of this article can be found under <https://doi.org/10.1002/sml.202504531>

DOI: 10.1002/sml.202504531

thickness of the semiconductor film (t) between the source and drain electrodes, which typically ranges from tens to hundreds of nanometers. Moreover, the channel area (A), which is expressed as $W \times d$ in the equation above, simply corresponds to the overlapped area of the source/drain electrodes, which typically ranges from several hundred to tens of thousands of square micrometers. Therefore, vECTs constructed by rearranging the device structure of conventional ECTs (cECTs) can exhibit higher g_m , which can then be described as $(A/t) \times \mu \times C^* \times (V_G - V_{th})$ in the saturation regime. By employing this geometric engineering, Facchetti et al. demonstrated that vECTs can achieve up to 206 times higher transconductance than cECTs when the same channel materials are used.^[22] Furthermore, they developed vertically stacked complementary logic circuits by stacking ECTs on top of each other. Their findings suggest that the new device structure not only improves the performance of ECTs but also increases the integration density of the devices.

To achieve high-performance vECTs, the channel materials must possess nanoscopic pores that facilitate ion penetration for volumetric charging and discharging. Therefore, solution-processed conjugated polymers have been the benchmark materials for ECT applications because they exhibit nanoscopic free volumes in thin-film states.^[1] Extensive side-chain engineering of these materials, which primarily affects ion-transport characteristics in thin films, has led to continuous improvements in ECT performance.^[34–39] However, most high-performance ECTs based on conjugated polymers exhibit p -type characteristics, wherein the hole current increases (decreases) when the films are doped (dedoped) with anions (cations).^[13,39,40] Promising alternative channel materials for ECTs with n -type characteristics include nanostructured inorganic materials, such as colloidal quantum dots (QDs). Thin films densely packed with QDs possess nanoscopic pores even in an ideally close-packed structure (the ideal packing fraction for spheres is 74%).^[41] Additionally, the organic ligands attached to the surface of the inorganic core create additional free volumes, further facilitating ion penetration. In fact, extensive fundamental studies using spectroelectrochemical methods have investigated how the discrete energy levels of different QD films are filled/emptied upon volumetric doping/dedoping processes with ions.^[42–47] However, in more practical device-level studies based on this principle,^[48–56] there is no clear explanation as to whether these devices operate through volumetric charging and discharging, as in ECTs, or through interfacial charging and discharging, as in EDLTs. Even though QD thin films are inherently structured to allow sufficient ion penetration, their operation has been interpreted using the EDLT mechanism in some studies.^[50,53,54] In these cases, the interpretation has been based on areal specific capacitance ($F\text{ cm}^{-2}$) rather than volumetric specific capacitance ($F\text{ cm}^{-3}$), suggesting a disconnect between the physical understanding of electrochemical charging in QD films and the device-level understanding of transistor operation, despite both being rooted in fundamentally similar principles. Furthermore, there have been no reported cases of using QD thin films as the channel of vECTs. Therefore, addressing this issue could create new possibilities for advancing high-performance transistor technologies.

Herein, we report the development of vertical QD ECTs (vQECTs) based on n -type InAs QD films. The relationship between the volume of the InAs QD films and their transduc-

tance and capacitance was examined through a systematic investigation, which revealed that the entire film exhibits volumetric capacitance rather than surface capacitance. This confirms that QD-based electrolyte-gated transistors operate through volumetric charging, following the mechanism of ECTs rather than that of EDLTs. Furthermore, by integrating n -type vQECTs with p -type vertical organic ECTs (vOECTs), we were able to fabricate vertically stacked hybrid inverters. These findings establish QD-based ECTs as viable candidates for high-performance vertical transistor architectures and compact logic circuits.

2. Results and Discussion

Figure 1a illustrates the structure of a vQECT based on a channel of n -type InAs QDs, which are one of the promising RoHS-compliant QD systems. (The material also exhibits a narrow bandgap yielding characteristic photo-response in the infrared regime.^[57]) The vQECTs operate based on the volumetric charging and discharging of the channel with cations of an ion gel electrolyte. The fabrication process is schematically illustrated in **Figure 1b**. Briefly, a Cr/Au contact was formed on a Si/SiO₂ substrate via thermal evaporation through a shadow mask to serve as the source electrode of the device. The InAs QD film was then spin-coated onto the substrate using an InAs QD dispersion (in toluene) containing photo-cross-linking agents, namely, 6-light-induced ligand cross-linkers (6-LiXers) [2-(((4-azido-2,3,5,6-tetrafluorobenzoyl)oxy)methyl)-2-((4-azido-2,3,5,6-tetrafluorobenzoyl)oxy)-2,2-bis(((4-azido-2,3,5,6-tetrafluorobenzoyl)oxy)methyl)propoxy)methyl)methyl]propane-1,3-diylbis(4-azido-2,3,5,6-tetrafluorobenzoate)]. Under UV illumination, the photo-cross-linking agent can form cross-links between the ligands of neighboring InAs QDs, modifying the solubility of the InAs QD film.^[58] The InAs QD film was photopatterned by selectively exposing it through a shadow mask and removing its uncross-linked regime using toluene. The patterned InAs QD films were treated with 1,2-ethanedithiol (EDT) to replace the insulating native ligands (oleic acid) and the cross-linkers, leading to enhanced electronic coupling between neighboring QDs.^[59] Note that both the patterned InAs QD film and the pristine InAs QD film without 6-LiXers exhibited similar degrees of ligand exchange after EDT treatment (93% and 95%, respectively) (**Figure S1**, Supporting Information). Additionally, the electrical conductivity of the patterned InAs QD films was also similar to that of the pristine InAs QD films without 6-LiXers (**Figure S2**, Supporting Information). These results confirm that the photo-cross-linkers did not alter the electrical transport characteristics of the EDT-treated InAs QD film since they were mostly removed during the EDT treatment. ZnMgO nanoparticles (NPs) ligated with butyl amine were subsequently photopatterned on top of the EDT-treated InAs QD patterns, which prevented the potential electrical leakage through the InAs QD film caused by metal penetration during top electrode deposition. vQECTs without ZnMgO NP layers exhibited drain current flow with no significant modulation at the 10^{-2} A level, indicating of electrical leakage (**Figure S3**, Supporting Information). For this purpose, here, the ZnMgO NPs were dispersed in chloroform and mixed with other photo-cross-linking agent, namely, 2-LiXers [ethane-1,2-diyl bis(4-azido-2,3,5,6-tetrafluorobenzoate)] (**Figure S4**, Supporting Information). The dispersion was then

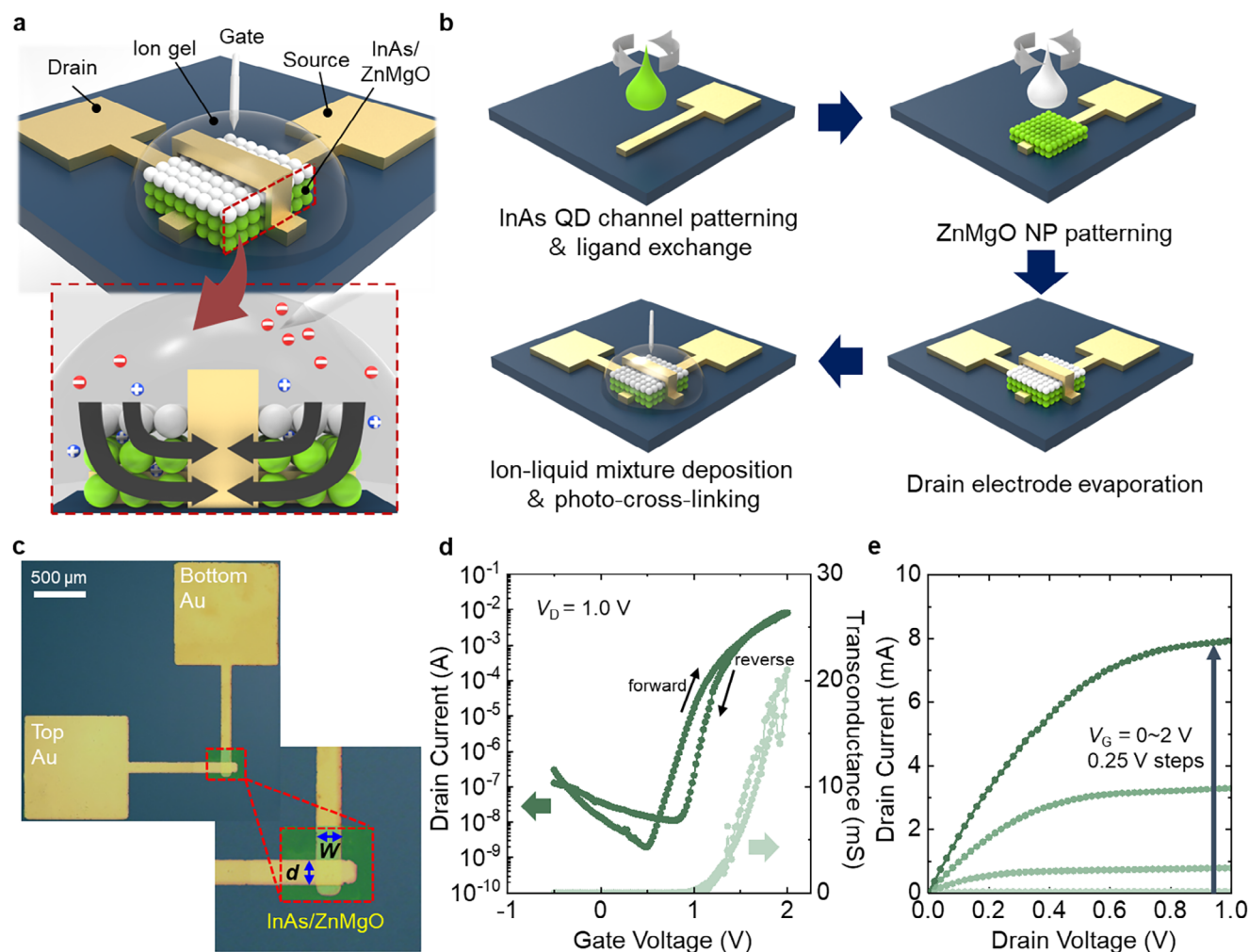


Figure 1. a). Schematic description of the structure and operation principles of vQECTs. b) Schematic description of the process for fabricating the vQECTs using photopatterned InAs QDs and ZnMgO NPs. c) Optical microscopy image, d) transfer characteristics, and e) output characteristics of the vQECTs.

spin-coated, and the resulting film was selectively exposed to UV light through a shadow mask, which induced cross-links between the organic ligands (butylamine) of the neighboring ZnMgO NPs. Subsequently, a development step was performed using fresh chloroform. Thereafter, an Au contact was deposited via thermal evaporation with a shadow mask to serve as the drain electrode of the device. This electrode overlapped the InAs QD/ZnMgO NP films in a direction rotated 90° from the bottom electrode. Finally, the ion-gel precursor solution, which comprised a mixture of an ionic liquid [1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide ([EMIM][TFSI])], a prepolymer [poly(ethylene glycol)diacrylate (PEG-DA)], and a photoinitiator [2-hydroxy-2-methylpropiophenone (HOMPP)], was dropped onto the regimes where the source and drain electrodes overlapped with the InAs QD/ZnMgO NP films. Upon 365 nm UV irradiation, an ion gel layer was formed through the cross-linking of PEG-DA that was photoinitiated with HOMPP.^[60] Finally, a gold tungsten tip coated with Ag/AgCl paste was contacted to the ion gel which served as the gate electrode. The overlapping area between the source and drain electrodes determined the trans-

port channel area [$A = (40 \times 40) - (100 \times 100) \mu\text{m}^2$] (Figure 1c). The length of the channel was determined from the thickness of the InAs QD/ZnMgO NP films (88 nm) sandwiched between the electrodes.

When a positive voltage was applied to the Ag/AgCl gate electrode connected to the ion gel layer, ionic polarization drove [EMIM]⁺ into the InAs QD/ZnMgO NP films, enabling the entire film to be charged with electrons (Figure 1a). When a negative voltage was subsequently applied to the gate electrode, the penetrated [EMIM]⁺ was extracted from the channel film, leading to the discharge of accumulated electrons. Consequently, sweeping V_G while applying a constant V_D (1.0 V) between the top and bottom electrodes induced a change in the channel current (I_D), which is a transfer characteristic of a transistor (Figure 1d left). This indicates that the device exhibits typical *n*-channel behavior, consistent with the *n*-type nature of the InAs QD/ZnMgO NP films. Note that the slight increase in I_D at more negative V_G s is attributed to the gate leakage current in the off state. Rather than weak *p*-channel behavior (Figure S5, Supporting Information). An I_D as high as 8 mA was attained at $V_G = 2$ V, a current

level that is rarely attainable in conventional *n*-type ECTs structures. Note that vECTs based solely on a photopatterned ZnMgO NP film channel—with a thickness comparable to that of the InAs QD/ZnMgO NP bilayer film—also exhibited *n*-channel behavior. However, both the degree of current modulation and the current level at $V_G = 2$ V were significantly lower than those observed in devices featuring the InAs QD/ZnMgO NP channel (Figure S6, Supporting Information). These results indicate that the underlying InAs QD film functions as the primary channel layer responsible for the high drain current. Figure 1d right presents the transconductance curve, which was extracted from the transfer characteristics using $g_m = \partial I_D / \partial V_G$. A maximum transconductance ($g_{m, \max}$) of 20.96 ± 2.16 mS was obtained. A finite hysteresis in transfer characteristics was observed between the forward and reverse scans of the vQECTs. Interestingly, the direction of the hysteresis depended on the scan rate. At a high scan rate above 0.15 V s⁻¹, the device displayed a typical anticlockwise hysteresis (Figure S7, Supporting Information), which is commonly observed in *n*-type ECTs. Such hysteresis is typically due to the sluggish motions of ions that penetrate the channel but are unable to promptly exit the channel during the fast turn off V_G scan. Conversely, at a slow scan rate of 0.08 V s⁻¹, a clockwise hysteresis was obtained. This clockwise hysteresis is attributable to the negation of the sluggish ionic effect at such a low scan rate, which instead reveals the electron-trapping effect possibly at the InAs QD surface (a clockwise hysteresis is typically obtained for *n*-channel transistors when the electron-trapping effect is prominent).^[61] Note that such hysteresis leads to a shift in V_{th} depending on the operation conditions of the device. When conventional logic circuits—where precise voltage control is essential for deterministic logic switching—are implemented using these transistors, the hysteresis can pose challenges including reduced noise margins and potential timing instability.^[62] However, this behavior may also be leveraged in alternative circuit architectures, such as memory devices or neuromorphic systems, where history-dependent responses and charge retention are beneficial.^[27] Figure 1e shows a series of output characteristics, I_D versus V_D relations, obtained from the vQECTs at different fixed V_G s (0 – 2 V, at 0.25 V steps). The shape of the output characteristics also follows typical *n*-channel behavior.

Prior to understanding the characteristics of the vQECTs in detail, we will discuss the critical role of ZnMgO NP layers. As mentioned above, vQECTs without ZnMgO NP layers exhibited significant electrical leakage (Figure S3, Supporting Information). We conjecture that this phenomenon was caused by microcracks in the InAs QD film, which were formed when the film was treated with EDT.^[59] In the presence of microcracks, the top electrode material may penetrate the crack during the deposition step, leading to an electrical shortage pathway. Despite attempts to fill cracks using repeated InAs QD deposition or EDT treatment conditions, reproducibility across devices was poor, prompting us to adopt an overlayer, which significantly improved fabrication consistency. We selected ZnMgO NP films for this purpose. ZnMgO NPs are well-known *n*-type materials used in QD devices.^[63] Despite the small conduction band minimum offset between the InAs QD and ZnMgO NP layers (Figure S8, Supporting Information), interfacial electron transport from InAs QD to ZnMgO layer – and eventually to the top drain electrode – should be feasible, particularly under a positive V_D ($= 1$ V).

Similar to QDs, ZnMgO NPs can also be photopatterned by cross-linking the ligands attached to their surface; however, this process does not require an additional ligand removal/replacement step. Therefore, the probability of microcracks forming in the NP film is minimal. Consequently, vQECTs with ZnMgO NP layers operated without exhibiting electrical shorts, as shown in Figure 1d,e.

To further investigate the role of the ZnMgO NP layer, we conducted additional experiment using two testbed devices with a cQECT structure, wherein the source/drain electrodes and the channel are arranged laterally, while the ion gel layer is laminated on top of the channel (Figure S9, Supporting Information): Testbed 1 comprised cQECTs with an InAs QD film channel placed underneath a ZnMgO NP film and Testbed 2 comprised cQECTs with only an InAs QD film channel. Here, all the InAs QD and ZnMgO NP films were deposited following the same protocol used to fabricate vQECTs shown in Figure 1. The *n*-type transfer curve of Testbed 2 indicates that InAs QD films are easily charged/discharged with [EMIM]⁺ (Figure S9, Supporting Information). The slightly higher drain current flow in Testbed 1 compared to Testbed 2 indicates that the top ZnMgO NP layer can additionally be charged/discharged with [EMIM]⁺. Overall, we conjecture that the ZnMgO NP layer in the vQECTs enables effective delivery of electrons from the InAs QD channel to the drain electrode, while also preventing electrical shorts between the source/drain electrodes.

Testbed 2 was also employed to validate the volumetric charging of the InAs QD films. We hypothesized that if ions can penetrate the nanoscopic pores within the InAs QD films and electrochemically charge the entire volume of the film, the current level of the device should scale with the thickness of the channel. In contrast, if 2D charging occurs locally at the QD film/ion gel interface, the channel current should remain nearly constant regardless of the channel thickness variation. Figure 2a shows the transfer characteristics of the cQECTs with different QD film thicknesses (46, 78, and 105 nm) obtained at $V_D = 1.0$ V. The morphology of these films examined through atomic force microscope (AFM) is shown in Figure 2b. The similar surface roughness of the InAs QD films suggests that any discrepancy in the electrical measurement can be attributed to the thickness variations. Clearly, the current levels and the corresponding g_m values consistently increased as channel thickness increased (Figure 2c). Figure 2d depicts the $g_{m, \max}$ values of these films as functions of $W \times t/L$ (where W and L are the channel width and length, as defined above for the cECT equation, respectively, and t is the film thickness). W and L were fixed to 1600 and 200 μ m, respectively. Clearly, the $g_{m, \max}$ of the device exhibited a linear relationship with t ($g_{m, \max} = (31.12 \pm 1.87) \frac{W}{L} t$, $R^2 = 0.9929$), indicating that the entire volume of the InAs QD film participates in electrical conduction and that the films are therefore charged volumetrically through electrolyte gating.

The V_D -dependent variation in the shape of the transfer characteristics provides additional evidence for the volumetric charging of InAs QD films. Figure 2e shows the transfer characteristics of cQECTs (the channel thickness = 46 nm) collected at two different V_D s (1.0 and 0.1 V). Unlike the transfer characteristics measured at $V_D = 1.0$ V, where I_D consistently increased as V_G increased, those measured at low $V_D = 0.1$ V displayed

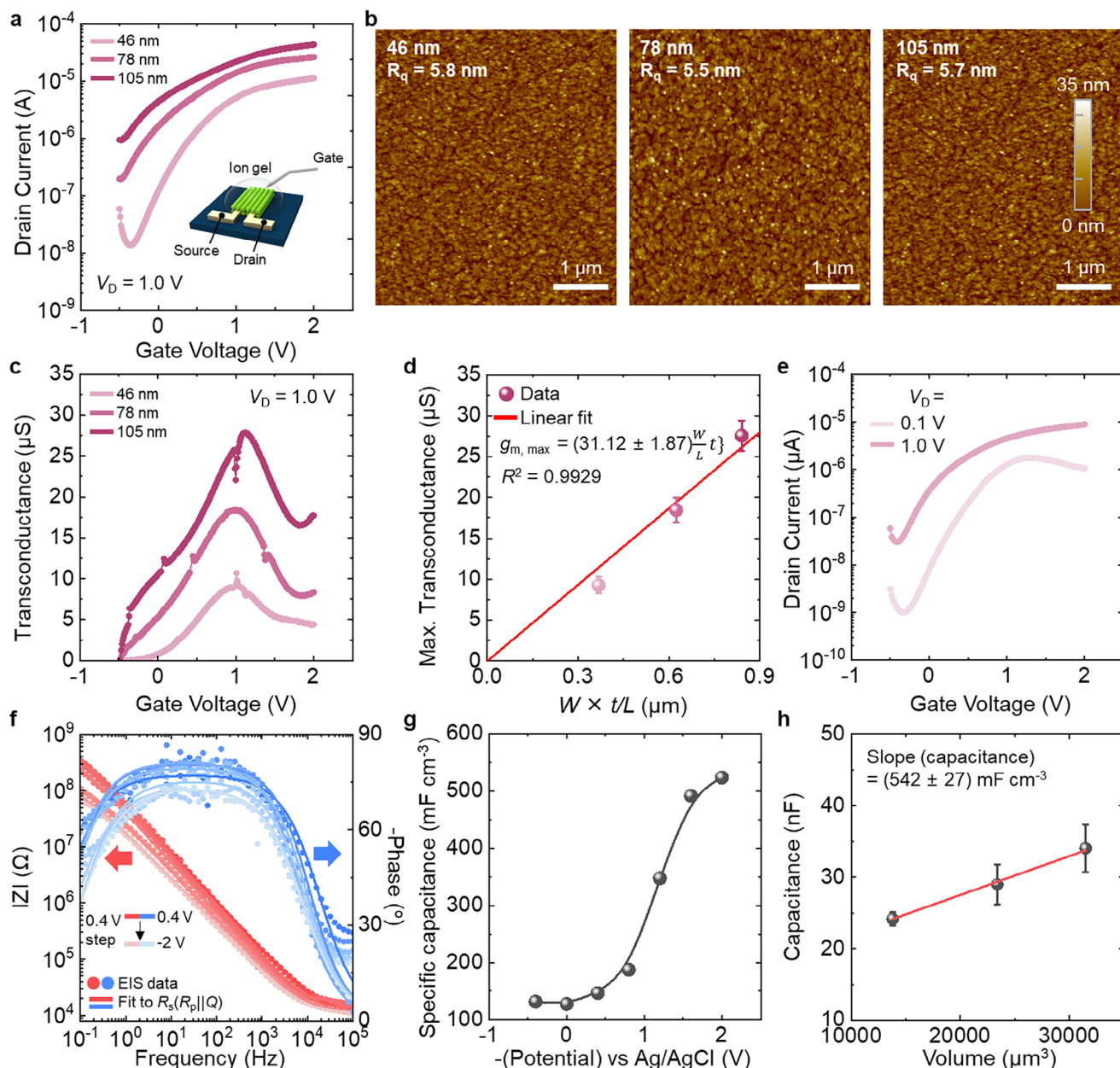


Figure 2. a). Transfer characteristics of the cQECTs with different InAs QD film thicknesses ($t = 46, 78$, and 105 nm). b) AFM images ($5 \mu\text{m} \times 5 \mu\text{m}$) of the InAs QD films with different thicknesses ($t = 46, 78$, and 105 nm). c) Transconductance curves of the cQECTs with different channel thicknesses ($t = 46, 78$, and 105 nm). d) Maximum transconductance of the cQECTs plotted as a function of channel geometry factors. e) Transfer characteristics of the cQECTs at different V_{DS} (0.1 and 1.0 V). f) Bode plot of the electrical impedance spectroscopy data for InAs QD films collected at different offset voltages ranging from 0.4 to -2.0 V in 0.4 V steps. The complex data were fitted to $R_s(R_p||Q)$. g) Volumetric specific capacitance of the InAs QD films as a function of the applied potential. h) Total capacitance of the InAs QD films as a function of the film volume.

a negative transconductance regime at V_G levels above 1.2 V, where I_D decreased as V_G increased. The output characteristics of these devices (Figure S10, Supporting Information) consistently demonstrated an unusual current level flip at low V_{DS} . Such an unusual negative transconductance at low V_{DS} and high V_G levels has also been observed in certain n -type organic ECTs (OECTs).^[6,8,38,64] This phenomenon is attributed to a substantial shift in the Fermi level of the channel caused by volumetric electrochemical doping, which drastically increases the injection bar-

rier at the source/channel heterojunction, leading to yield non-ohmic charge injection.^[38] We believe that the same explanation can be applied to our findings. Owing to the substantial volumetric charging of InAs QD films induced by electrochemical doping, the Fermi level of the channel rises substantially, resulting in the formation of a sufficiently high injection barrier at the source/channel junction. Although this injection barrier does not have a substantial impact under high V_{DS} , it can noticeably suppress electron injection under low V_{DS} , leading to the generation

of a negative transconductance regime at high V_G s (Figure S11, Supporting Information). Overall, the negative transconductance regime observed in our cQECTs under low- V_D and high- V_G operation conditions further indicates that the InAs QD films are volumetrically charged during the operation.

Furthermore, we performed electrochemical impedance spectroscopy (EIS) to investigate the volumetric capacitance of the InAs QD films. For the EIS measurements, we employed a metal–electrolyte–semiconductor structure. The InAs QD film, which served as the semiconductor layer, was deposited on a Au electrode (0.03 cm^2) serving as the working electrode. The ion gel layer, which served as the electrolyte layer, was cast onto the InAs QD film, and an Ag/AgCl tip was used as the reference and counter electrode (Figure S12, Supporting Information). Figure 2f shows the Bode plot obtained from 10^5 to 0.1 Hz with an alternating current amplitude of 10 mV at offset voltage ranging from 0.4 to -2.0 V in 0.4 V steps (the offset voltage of -2 V corresponds to a condition where $+2 \text{ V}$ is applied to the Ag/AgCl tip relative to the Au electrode). This data was well-fitted to the Randles circuit model $R_s(R_p||Q)$, where R_s is the electrical resistance of the electrolyte, R_p is the polarization resistance, and Q is the constant phase element. The capacitance spectra were extracted using an equation $C = 1/(2\pi f \times \text{Im}(Z))$, where f is the frequency and Z is the complex impedance (Figure S12, Supporting Information).^[65] Next, the volumetric specific capacitance of the InAs QD films was estimated by dividing the capacitance measured at 1 Hz by the volume of the films. The estimated values were plotted as a function of the applied potential (Figure 2g). Notably, the specific capacitance increased as the potential increased, and it reached up to 523 mF cm^{-3} at 2 V , indicating that volumetric charging was enhanced as more $[\text{EMIM}]^+$ penetrated the InAs QD at increased potentials. We also plotted the measured total capacitance values (in F) at 2 V for the InAs QD films with different thicknesses as a function of film volume (Figure 2h). Clearly, the capacitance increased as the film volume increased. The volumetric specific capacitance obtained from the slope of this plot was $542 \pm 27 \text{ mF cm}^{-3}$, which is consistent with the values obtained from Figure 2g at 2 V . Collectively, these results indicate that the InAs QD films are volumetrically charged by ions, allowing the entire film to be electrochemically doped.

After confirming the volumetric charging capability of the InAs QD films, we investigated the geometry-dependent performance of vQECT devices. Specifically, we varied the channel area of the device while keeping the thickness of the ZnMgO NP and InAs QD films constant (88 nm). Figure 3a shows the transfer characteristics of the vQECTs with three different channel areas [$A = 40 \times 40$, 70×70 , and $100 \times 100 \mu\text{m}^2$]. The vQECTs with the largest channel area exhibited the highest on-current (I_D^{on} , defined as the drain current at $V_G = 2 \text{ V}$) and $g_{m, \text{max}}$. Figure 3b shows plots of I_D^{on} and $g_{m, \text{max}}$ as functions of A . Although the vertical transistor equation ($I_D = (A/2t) \times \mu \times C^* \times (V_G - V_{\text{th}})^2$) and its corresponding maximum transconductance ($g_{m, \text{max}} = (A/t) \times \mu \times C^* \times (V_G - V_{\text{th}})$) predict that I_D and $g_{m, \text{max}}$ should exhibit simple linear scaling relations with A , a sublinear relation was found in this study. This can be explained by the extended distance that the ions (originally located outside the film at the off state) must travel to fully charge the InAs QD films between the electrodes when A is relatively large. In fact, the turn-on voltage

(V_{on}), which represents the minimum gate voltage required to electrochemically charge the InAs QD films, also exhibited geometry dependence. A more positive V_{on} was obtained at large A values, indicating that a high gate voltage is required to drive ions into the InAs QD film between the electrodes with A is large.

In terms of device performance, our vQECTs exhibited exceptionally high I_D^{on} for a specified channel area. In fact, the device with $A = 100 \times 100 \mu\text{m}^2$ exhibited an on-current density (J_D^{on}) of $79.5 \pm 3.54 \text{ A cm}^{-2}$, which is by far the highest integration area normalized current ($I_{D,A}^{\text{on}}$, i.e., drain current divided by the projected channel area on the substrate) value reported for n -channel transistors based on ionic liquid gating.^[28,31,54–56,66–72] This metric, introduced from Faccetti et al.^[22] is particularly relevant for vECTs, as it reflects their advantage in terms of areal integration. In the case of vertical devices, $I_{D,A}^{\text{on}}$ coincides with J_D^{on} , while in planar devices the two metrics differ due to the lateral charge transport geometry. It is worth noting that our vQECTs not only exhibited the highest $I_{D,A}^{\text{on}}$ value but also achieved the highest on/off current ratio ($(4.3 \pm 0.22) \times 10^6$), which is a crucial indicator of the switching capability of a transistor (Figure 3c).

In addition to the excellent switching characteristics, the vQECTs also exhibited decent stability characteristics. Figure 3d shows the bias stability of the transfer characteristics under a constant V_G (2 V) for different periods (0 – 2400 sec). The device parameters— $I_{D,A}^{\text{on}}$, on/off current ratio, and subthreshold swing (V_{ss})—remained nearly unchanged under these test conditions (Figure 3e). Figure 3f shows some of the transfer characteristics of the vQECTs that were measured at 7-day intervals while being stored in an N_2 -filled glove box. While I_D slightly decreased over time, the on/off current ratio and the V_{ss} remained nearly consistent, indicating that the device exhibited strong stability under the N_2 -storage conditions (Figure 3g). Moreover, the operational stability of the device is demonstrated in Figure 3h, which shows the drain current response obtained when square-wave-formed input signals (i.e., $V_G = 2$ and 0 V) were applied. The vQECTs exhibited reliable characteristics over more than $10\,000$ switching cycles, a feature that has not been achieved by previously reported n -channel ionic liquid-gated transistors. Although the same level of stability was not observed under ambient conditions, the confirmed intrinsic stability under N_2 storage suggests that, with proper encapsulation or environmental protection strategies, the vQECTs hold potential for practical applications.

Finally, a vertically stacked complementary inverter was fabricated to demonstrate the feasibility of vQECTs in vertical logic applications. The fabrication of a complementary inverter comprising an n -channel transistor and a p -channel transistor is regarded as a primary demonstration toward fabricating more complicated logic gates. To fabricate the inverter, we employed a p -type vQECT based on benchmark poly(3-hexylthiophene-2,5-diyl) (P3HT) as a complementary channel to our n -type vQECT (Figure 4a).^[27] Specifically, we formed the n -type vQECT on top of a bottom Au contact, inserted a middle Au contact, and then stacked the p -type vQECT channel on top together with a top Au contact. Finally, an ion gel was placed on the vertical channels to gate both the p -channel and n -channel of the inverter (Figure S1, Supporting Information). For the inverter operation, the bottom

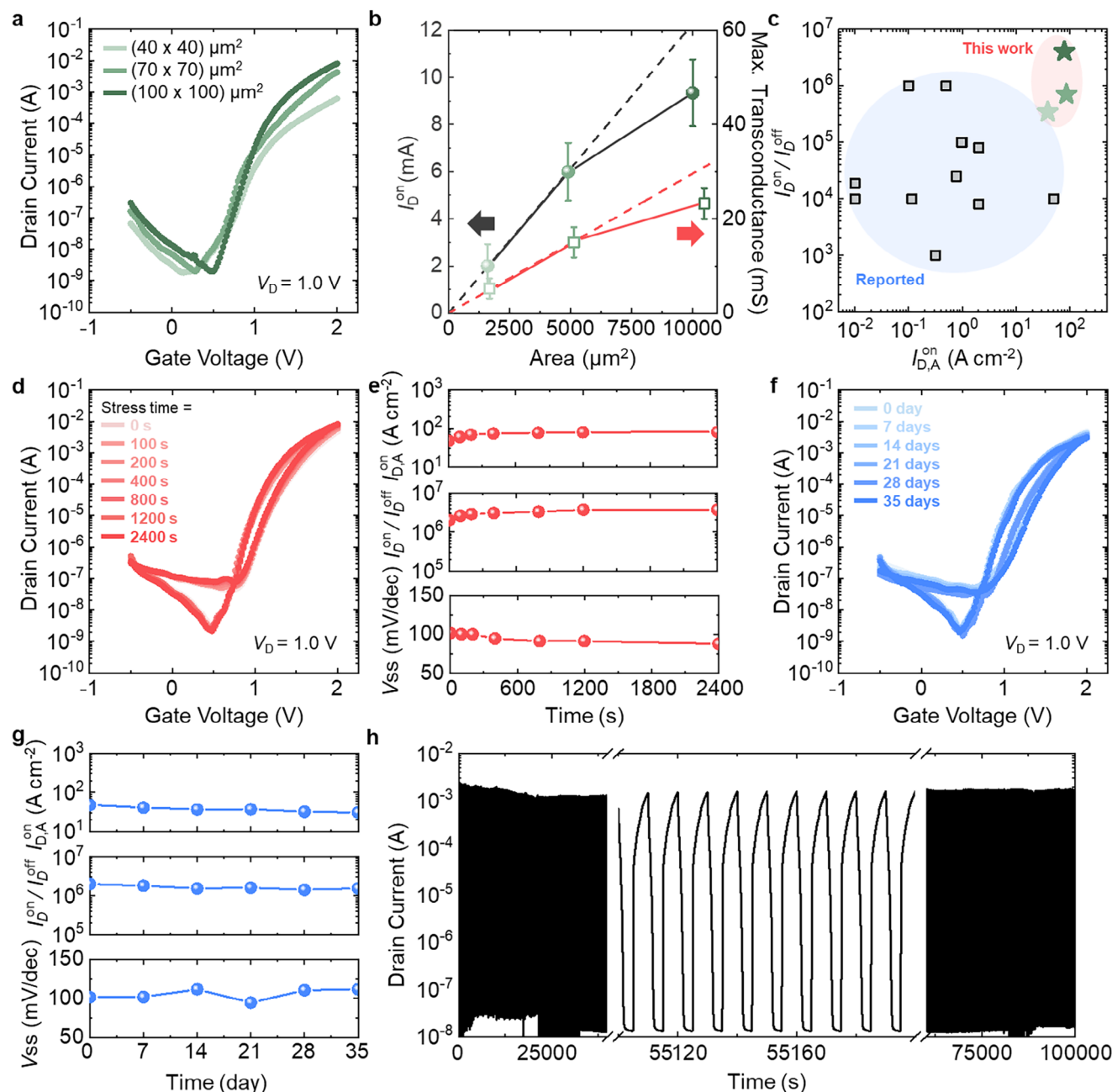


Figure 3. a). Transfer characteristics of vQECTs with different channel areas. b). Averaged I_D^{on} (left y-axis) and averaged $g_{m, \text{max}}$ (right y-axis) of the vQECTs with different channel areas. c). Comparison of on/off current ratio- $I_{D, A}^{\text{on}}$ for n-type ionic liquid-gated transistors. The Sources are detailed in Table S1 (Supporting Information). d). Transfer characteristics and e) parameters ($I_{D, A}^{\text{on}} / I_{D, A}^{\text{off}}$, on/off current ratio, and V_{ss}) of the vQECTs under a constant V_G for 2400 s. f). Transfer characteristics and g) parameters ($I_{D, A}^{\text{on}} / I_{D, A}^{\text{off}}$, on/off current ratio, and V_{ss}) of the vQECTs stored inside an N_2 -filled glove box for 35 days. h). Operational stability of the vQECTs over 100 000 s at $V_D = 1.0 \text{ V}$ and V_G switching between 0 and 2 V.

electrode functioned as the ground (GND) electrode, the middle electrode served as the output voltage (V_{OUT}) readout, the top electrode served as the supply voltage (V_{DD}) electrode, and the Ag/AgCl tip contacting the ion gel served as the contact for input voltage (V_{IN}) (Figure 4a). As shown in Figure 4b, all electrodes were carefully aligned to overlap the channel areas of the bottom vQECTs and top vQECTs. The transfer characteristics of the p-type vQECTs were balanced with those of the n-type vQECTs

(Figure 4c), causing the hybrid inverters to exhibit a typical inversion response. The V_{OUT} versus V_{IN} relations were collected at different V_{DD} s ($V_{\text{DD}} = 0.2\text{--}1.0 \text{ V}$, 0.2 V steps), as shown in Figure 4d. A "1" output logic state ($V_{\text{OUT}} = V_{\text{DD}}$) appeared at $V_{\text{IN}} = 0 \text{ V}$ (where the n-type vQECTs were turned off and the p-type vQECTs were turned on), and a "0" output logic state ($V_{\text{OUT}} = 0 \text{ V}$) appeared at $V_{\text{IN}} = 2 \text{ V}$ (where the n-type vQECTs were on and the p-type vQECTs were off). The signal gain (defined as $\partial V_{\text{OUT}} / \partial V_{\text{IN}}$)

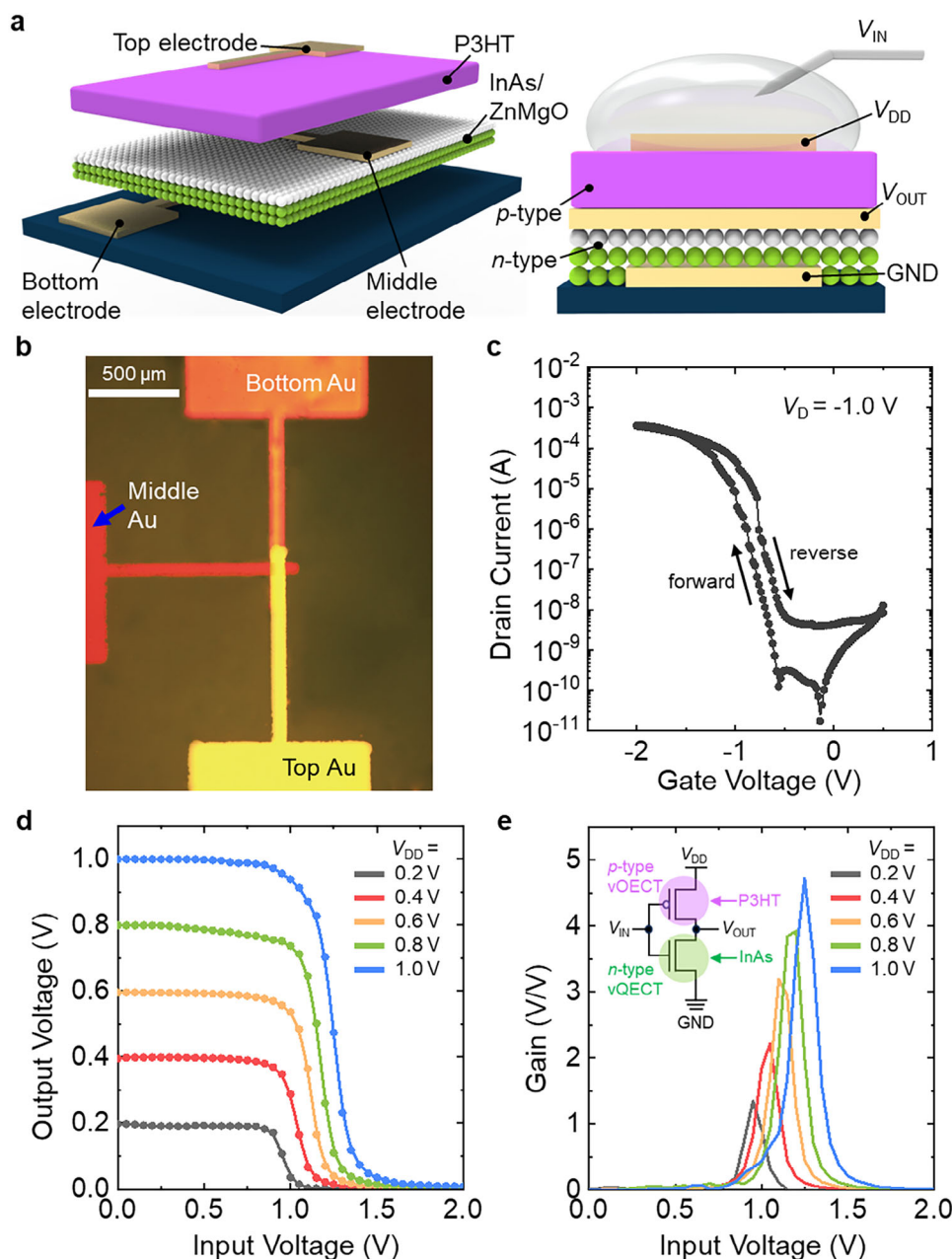


Figure 4. a). Schematic description of hybrid inverters (left) with cross-sectional illustrations (right). b) Optical microscopy image of hybrid inverters. c) Transfer characteristics of the P3HT-based vOECTs. d) Voltage-transfer characteristics and e) inverter gain signal of the hybrid inverters.

of hybrid inverters was determined as ≈ 4.7 at $V_{DD} = 1$ V (Figure 4e).

3. Conclusion

In this study, we implemented vQECTs using *n*-type InAs QD thin films. Our systematic investigation confirmed that the InAs QD films exhibit volumetric capacitance, validating their operation as true ECTs rather than EDLTs. The vQECTs exhibited high transconductance ($g_{m, \max} = 20.96 \pm 2.16$ mS) and a record-high $I_{D,A}^{on} = 79.5 \pm 3.54$ A cm $^{-2}$ among *n*-type ionic liquid-

gated transistors, along with excellent operational stability. Furthermore, by integrating *n*-type vQECTs with *p*-type vOECTs, we successfully fabricated vertically stacked hybrid inverters, which achieved a signal gain of ~ 4.7 . These results highlight the potential of QD-based ECTs for use in high-performance vertical transistor architectures. Our study provides a new platform for developing compact, high-density electronic devices and creates avenues for constructing next-generation electrolyte-gated logic circuits. Future work will focus on optimizing material properties and exploring additional device architectures to further enhance performance.

4. Experimental Section

Chemicals: Indium acetate (99.999%), oleic acid ($\geq 99\%$), and 1-octadecene (ODE; 99.999%) were purchased from UNIAM. Dioctylamine ($\geq 97\%$), hexane (anhydrous, 95%), 1-butanol (anhydrous, 99.8%), and toluene (anhydrous, 99.8%) were purchased from Sigma-Aldrich. Trimethylsilylarsine ((TMSi)₃As; 99%) was purchased from JSI Silicone. Additionally, 2-LiXer and 6-LiXer were prepared according to the protocol described previously.^[58,73] ZnMgO NPs were prepared using previously reported methods with butylamine instead of ethanolamine in the final step.^[74]

Materials for Device Fabrication: Si substrates (n^{++} , $< 0.005 \Omega$) with a thermally grown SiO₂ layer (300 nm) were purchased from Namkang Hi-Tech. [EMIM][TFSI] was purchased from C-TRI Korea. EDT (98%), PEG-DA (average M_n 575), HOMPP (97%), and P3HT (average M_w = 20 000–45 000) were purchased from Sigma-Aldrich.

InAs QD Synthesis: The QDs were synthesized using previously reported methods with minor modifications.^[75] To synthesize InAs seed QDs, an indium oleate solution was prepared by mixing 0.29 g (1 mmol) of indium acetate with 0.85 g (3 mmol) of oleic acid in 5 mL of ODE and degassing the mixture in a three-neck flask under a vacuum at 100 °C for 2 h. The As solution was then prepared by mixing 0.14 g (0.5 mmol) of (TMSi)₃As with 0.36 g (1.5 mmol) of dioctylamine dissolved in 1 mL of degassed ODE solution, and the solution was stored inside a nitrogen-filled glove box at 60 °C for 30 min. The degassed indium oleate solution was heated to 300 °C under nitrogen conditions. The prepared As solution was rapidly injected into the indium oleate solution at 300 °C.

To synthesize amorphous InAs nanoclusters, an indium oleate solution was prepared by mixing 1.74 g (6 mmol) of indium acetate with 5.10 g (18 mmol) of oleic acid in 30 mL of ODE and degassing the mixture at 100 °C for 2 h under vacuum. Thereafter, the As solution was prepared by mixing 0.84 g (3 mmol) of (TMSi)₃As with 2.17 g (9 mmol) of dioctylamine dissolved in 6 mL of degassed ODE solution, and the solution was stored inside a nitrogen-filled glove box at 60 °C for 30 min. The As solution was then injected into the indium oleate solution at room temperature and stirred for 10 min.

To synthesize the InAs QDs, the InAs nanocluster solution was continuously injected into the InAs seed QD solution through a syringe pump at a growth temperature of 300 °C at desired rates. After the growth reaction was terminated, the InAs QDs were repeatedly purified using butanol and hexane. Finally, the synthesized InAs QDs were dispersed in toluene.

Device Fabrication: To fabricate the vQECTs, the Si/SiO₂ substrates were first cleaned sequentially with acetone, 2-propanol, and distilled water. Next, the Cr/Au electrode was thermally evaporated onto the substrates at deposition rates of 0.1/1.0 Å s⁻¹, respectively. Subsequently, dispersions (20 mg mL⁻¹ in toluene) of InAs QDs mixed with 6-LiXer (1 wt.% relative to the weight of the InAs QDs) were spin-coated (2000 rpm, 30 s) onto the substrates. The channel regimes of the vQECTs were selectively irradiated with UV light (254 nm, 0.4 mW cm⁻², corresponding exposure: 2 mJ cm⁻²) using a shadow mask for 10 s. The uncross-linked regime of the InAs QD film was removed using pure toluene in order to form the patterns. For ligand exchange, the InAs QD films were treated with 50 mM EDT in acetonitrile for a dwelling time of 2 min and then rinsed with pure acetonitrile. The channel-patterning to ligand-exchange steps were repeated once more. Thereafter, dispersions (20 mg mL⁻¹ in chloroform) of ZnMgO NPs mixed with 2-LiXer (5 wt.% relative to the weight of the ZnMgO NPs) were spin-coated (2000 rpm, 60 s). The areas of ZnMgO NPs film having the InAs QD patterns underneath were selectively irradiated with UV light (254 nm) through the same shadow mask used to pattern the InAs QDs, and then patterned ZnMgO NP films were formed by removing the uncross-linked regime of the ZnMgO NP film using pure chloroform. The Au electrode was thermally evaporated onto the ZnMgO NP layer at a deposition rate of 1.0 Å s⁻¹. Finally, a liquid mixture of PEG-DA, HOMPP, and [EMIM][TFSI] (weight ratio = 2:1:22) was poured onto the regimes where the two electrodes and the InAs QD/ZnMgO NP films overlapped. The liquid mixture was subsequently exposed to UV light (365 nm, 0.4 mW cm⁻², corresponding exposure = 2 mJ cm⁻²) for 60 s. The detailed conditions of the fabrica-

tion process of the cQECTs, including InAs QD channel formation and ion gel deposition, were the same as those for the vQECTs. The entire fabrication process was performed under an inert atmosphere inside an N₂-filled glove box.

To fabricate the complementary inverter, the P3HT solution in chloroform (10 mg mL⁻¹) was prepared by stirring the mixture solution at 50 °C for 5 h. Next, 2-LiXer in chloroform (5 wt.% relative to the weight of the P3HT) was added to the P3HT solution. Subsequently, the 2-LiXer containing P3HT dispersions was spin-coated (2000 rpm, 60 s) onto the vQECTs. After being irradiated with UV light (254 nm, 0.4 mW cm⁻², corresponding exposure: 2 mJ cm⁻²) and rinsed with pure chloroform, the samples were dried for 12 h in an N₂-filled glove box. The Au electrode was thermally evaporated onto the P3HT layer at a deposition rate of 1.0 Å s⁻¹. Finally, a liquid mixture of PEG-DA, HOMPP, and [EMIM][TFSI] (weight ratio = 2:1:22) was poured onto the regimes where the three electrodes overlapped. The liquid mixture was then exposed to UV light (365 nm, 0.4 mW cm⁻², corresponding exposure 2 mJ cm⁻²) for 60 s.

Characterization: FT-IR spectra were collected using an FT-IR 4700 spectroscopy (JASCO) under ambient conditions. The samples for FT-IR analysis were prepared on bare Si substrates. The surface morphology and thickness of films were confirmed using a Park XE7 (Park Systems) under ambient conditions. Electrical measurements were performed on all the devices using a probe station (MSTECH M5VC) connected to a Keithley 4200-SCS parameter analyzer under vacuum conditions. EIS measurements were performed using a potentiostat (Metrohm MULTI AUTOLAB M204) connected to a probe station under vacuum conditions.

Statistical Analysis: All the average and standard deviation values are obtained from more than 10 samples.

Supporting Information

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

Acknowledgements

H. J. and S. P. contributed equally to this work. This work was supported by the National Research Foundation of Korea (NRF) grant funded by the Ministry of Science, ICT, and Future Planning (NRF-2019M3D1A1078299 and RS-2021-NR059263). This work was partially supported by the Samsung Research Funding and Incubation Center of Samsung Electronics (SRFC-MA1901-51).

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

colloidal InAs quantum dot, inverter, ion gel, photopatterning, vertical electrochemical transistors

Received: April 11, 2025
Revised: June 18, 2025
Published online: June 30, 2025

- [1] J. Rivnay, S. Inal, A. Salleo, R. M. Owens, M. Berggren, G. G. Malliaras, *Nat. Rev. Mater.* **2018**, 3, 17086.
- [2] J. T. Friedlein, R. R. McLeod, J. Rivnay, *Org. Electron.* **2018**, 63, 398.
- [3] H. Sun, J. Gerasimov, M. Berggren, S. Fabiano, *J. Mater. Chem. C* **2018**, 6, 11778.
- [4] H. Jo, W. Lee, H. Jung, D. M. Park, H. Lee, M. S. Kang, *ACS Appl. Mater. Interfaces* **2021**, 13, 13541.
- [5] Y. Yao, W. Huang, J. Chen, X. Liu, L. Bai, W. Chen, Y. Cheng, J. Ping, T. J. Marks, A. Facchetti, *Adv. Mater.* **2023**, 35, 2209906.
- [6] Y. Wang, E. Zeglio, L. Wang, S. Cong, G. Zhu, H. Liao, J. Duan, Y. Zhou, Z. Li, D. Mawad, A. Herland, W. Yue, I. McCulloch, *Adv. Funct. Mater.* **2022**, 32, 2111439.
- [7] K. Feng, W. Shan, J. Wang, J.-W. Lee, W. Yang, W. Wu, Y. Wang, B. J. Kim, X. Guo, H. Guo, *Adv. Mater.* **2022**, 34, 2201340.
- [8] I. P. Maria, S. Griggs, R. B. Rashid, B. D. Paulsen, J. Surgailis, K. Thorley, V. N. Le, G. T. Harrison, C. Corbue, R. Hallani, A. Giovannitti, A. F. Paterson, S. Inal, J. Rivnay, I. McCulloch, *Chem. Mater.* **2022**, 34, 8593.
- [9] J. Duan, G. Zhu, J. Chen, C. Zhang, X. Zhu, H. Liao, Z. Li, H. Hu, I. McCulloch, C. B. Nielsen, W. Yue, *Adv. Mater.* **2023**, 35, 2300252.
- [10] Y. Wang, G. Zhu, E. Zeglio, T. C. H. Castillo, S. Haseena, M. K. Ravva, S. Cong, J. Chen, L. Lan, Z. Li, A. Herland, I. McCulloch, S. Inal, W. Yue, *Chem. Mater.* **2023**, 35, 405.
- [11] X. Liu, Y. Xiao, C. Yan, P. Du, F. Zhang, H. Xin, *ACS Appl. Mater. Interfaces* **2025**, 17, 8072.
- [12] M. Moser, A. Savva, K. Thorley, B. D. Paulsen, T. C. Hidalgo, D. Ohayon, H. Chen, A. Giovannitti, A. Marks, N. Gasparini, A. Wadsworth, J. Rivnay, S. Inal, I. McCulloch, *Angew. Chem., Int. Ed.* **2021**, 60, 7777.
- [13] R. K. Hallani, B. D. Paulsen, A. J. Petty II, R. Sheelamantula, M. Moser, K. J. Thorley, W. Sohn, R. B. Rashid, A. Savva, S. Moro, J. P. Parker, O. Drury, M. Alsufyani, M. Neophytou, J. Kosco, S. Inal, G. Costantini, J. Rivnay, I. McCulloch, *J. Am. Chem. Soc.* **2021**, 143, 11007.
- [14] Y. Wang, A. Hamidi-Sakr, J. Surgailis, Y. Zhou, H. Liao, J. Chen, G. Zhu, Z. Li, S. Inal, W. Yue, *J. Mater. Chem. C* **2021**, 9, 13338.
- [15] J. So, T. Kim, J. Shin, D. Kim, F. S. Kim, *Macromol. Res.* **2023**, 31, 1095.
- [16] J. Rivnay, P. Leleux, M. Ferro, M. Sessolo, A. Williamson, D. A. Koutsouras, D. Khodagholy, M. Ramuz, X. Strakosas, R. M. Owens, C. Benar, J.-M. Badier, C. Bernard, G. G. Malliaras, *Sci. Adv.* **2015**, 1, 1400251.
- [17] P. C. Hütter, T. Rothländer, A. Haase, G. Trimmel, B. Stadlober, *Appl. Phys. Lett.* **2013**, 103, 043308.
- [18] S. Inal, G. G. Malliaras, J. Rivnay, *Nat. Commun.* **2017**, 8, 1767.
- [19] P. R. Paudel, V. Kaphle, D. Dahal, R. K. Radha Krishnan, B. Lüssem, *Adv. Funct. Mater.* **2021**, 31, 2004939.
- [20] G. D. Spyropoulos, J. N. Gelinis, D. Khodagholy, *Sci. Adv.* **2019**, 5, aau7378.
- [21] P. R. Paudel, M. Skowrons, D. Dahal, R. K. Radha Krishnan, B. Lüssem, *Adv. Theor. Simul.* **2022**, 5, 2100563.
- [22] W. Huang, J. Chen, Y. Yao, D. Zheng, X. Ji, L.-W. Feng, D. Moore, N. R. Glavin, M. Xie, Y. Chen, R. M. Pankow, A. Surendran, Z. Wang, Y. Xia, L. Bai, J. Rivnay, J. Ping, X. Guo, Y. Cheng, T. J. Marks, A. Facchetti, *Nature* **2023**, 613, 496.
- [23] J. Kim, R. M. Pankow, Y. Cho, I. D. Duplessis, F. Qin, D. Meli, R. Daso, D. Zheng, W. Huang, J. Rivnay, T. J. Marks, A. Facchetti, *Nat. Electron.* **2024**, 7, 234.
- [24] Y. Lai, J. Cheng, M. Xie, J. Chen, G. Zhu, W. Huang, L.-W. Feng, *Angew. Chem., Int. Ed.* **2024**, 63, 202401773.
- [25] Y. Choi, S. Oh, C. Qian, J.-H. Park, J. H. Cho, *Nat. Commun.* **2020**, 11, 4595.
- [26] J. W. Moon, D. U. Lim, Y. N. Kim, J. H. Kim, J. H. Kim, S. B. Jo, J. H. Cho, *Adv. Funct. Mater.* **2023**, 33, 2305440.
- [27] Y. Choi, D. H. Ho, S. Kim, Y. J. Choi, D. G. Roe, I. C. Kwak, J. Min, H. Han, W. Gao, J. H. Cho, *Sci. Adv.* **2023**, 9, adg5946.
- [28] S. Y. Jeong, J. W. Moon, S. Lee, Z. Wu, S. H. Park, J. H. Cho, H. Y. Woo, *Adv. Electron. Mater.* **2023**, 9, 2300053.
- [29] Y. Cho, L. Gao, Y. Yao, J. Kim, D. Zhang, G. Forti, I. Duplessis, Y. Wang, R. M. Pankow, X. Ji, J. Rivnay, T. J. Marks, A. Facchetti, *Angew. Chem., Int. Ed.* **2025**, 64, 202414180.
- [30] Z. Deng, Y. Yu, Y. Zhou, J. Zhou, M. Xie, B. Tao, Y. Lai, J. Wen, Z. Fan, X. Liu, D. Zhao, L.-W. Feng, Y. Cheng, C.-G. Huang, W. Yue, W. Huang, *Adv. Mater.* **2024**, 36, 2405115.
- [31] S. Y. Jeong, S. H. Park, J. Park, J. W. Moon, J. H. Cho, H. Y. Woo, *Dyes Pigm* **2024**, 228, 112201.
- [32] Z. Laswick, X. Wu, A. Surendran, Z. Zhou, X. Ji, G. M. Matrone, W. L. Leong, J. Rivnay, *Nat. Commun.* **2024**, 15, 6309.
- [33] S. Hou, W. Zuo, Q. Fang, P. Lu, B. Tao, M. Xie, G. Hu, J. Zhou, L.-W. Feng, W. Huang, *ACS Appl. Mater. Interfaces* **2025**, 17, 5176.
- [34] A. Giovannitti, D.-T. Sbircea, S. Inal, C. B. Nielsen, E. Bandiello, D. A. Hanifi, M. Sessolo, G. G. Malliaras, I. McCulloch, J. Rivnay, *Proc. Natl. Acad. Sci. U.S.A.* **2016**, 113, 12017.
- [35] D. Ohayon, A. Savva, W. Du, B. D. Paulsen, I. Uguz, R. S. Ashraf, J. Rivnay, I. McCulloch, S. Inal, *ACS Appl. Mater. Interfaces* **2021**, 13, 4253.
- [36] I. P. Maria, B. D. Paulsen, A. Savva, D. Ohayon, R. Wu, R. Hallani, A. Basu, W. Du, T. D. Anthopoulos, S. Inal, J. Rivnay, I. McCulloch, A. Giovannitti, *Adv. Funct. Mater.* **2021**, 31, 2008718.
- [37] K. Feng, W. Shan, S. Ma, Z. Wu, J. Chen, H. Guo, B. Liu, J. Wang, B. Li, H. Y. Woo, S. Fabiano, W. Huang, X. Guo, *Angew. Chem., Int. Ed.* **2021**, 60, 24198.
- [38] D. Jeong, I.-Y. Jo, S. Lee, J. H. Kim, Y. Kim, D. Kim, J. R. Reynolds, M.-H. Yoon, B. J. Kim, *Adv. Funct. Mater.* **2022**, 32, 2111950.
- [39] M. Moser, T. C. Hidalgo, J. Surgailis, J. Gladisch, S. Ghosh, R. Sheelamantula, Q. Thiburce, A. Giovannitti, A. Salleo, N. Gasparini, A. Wadsworth, I. Zozoulenko, M. Berggren, E. Stavrinidou, S. Inal, I. McCulloch, *Adv. Mater.* **2020**, 32, 2002748.
- [40] I.-Y. Jo, D. Jeong, Y. Moon, D. Lee, S. Lee, J.-G. Choi, D. Nam, J. H. Kim, J. Cho, S. Cho, D.-Y. Kim, H. Ahn, B. J. Kim, M.-H. Yoon, *Adv. Mater.* **2024**, 36, 2307402.
- [41] Y. Wu, Z. Fan, Y. Lu, *J. Mater. Sci.* **2003**, 38, 2019.
- [42] P. Hoyer, R. Eichberger, H. Weller, *Ber. Bunsenges. Phys. Chem.* **1993**, 97, 630.
- [43] A. L. Roest, A. J. Houtepen, J. J. Kelly, D. Vanmaekelbergh, *Faraday Discuss.* **2004**, 125, 55.
- [44] C. Wang, M. Shim, P. Guyot-Sionnest, *Science* **2001**, 291, 2390.
- [45] A. J. Houtepen, D. Vanmaekelbergh, *J. Phys. Chem. B* **2005**, 109, 19634.
- [46] S. Gudjonsdottir, W. van der Stam, N. Kirkwood, W. H. Evers, A. J. Houtepen, *J. Am. Chem. Soc.* **2018**, 140, 6582.
- [47] D. Vanmaekelbergh, A. J. Houtepen, J. J. Kelly, *Electrochim. Acta* **2007**, 53, 1140.
- [48] M. S. Kang, J. Lee, D. J. Norris, C. D. Frisbie, *Nano Lett.* **2009**, 9, 3848.
- [49] S. Z. Bisri, C. Piliago, M. Yarema, W. Heiss, M. A. Loi, *Adv. Mater.* **2013**, 25, 4309.
- [50] K. Shibata, H. Yuan, Y. Iwasa, K. Hirakawa, *Nat. Commun.* **2013**, 4, 2664.
- [51] D. M. Balazs, K. I. Bijlsma, H.-H. Fang, D. N. Dirin, M. Döbeli, M. V. Kovalenko, M. A. Loi, *Sci. Adv.* **2017**, 3, aao1558.
- [52] A. Sahu, M. S. Kang, A. Kompch, C. Notthoff, A. W. Wills, D. Deng, M. Winterer, C. D. Frisbie, D. J. Norris, *Nano Lett.* **2012**, 12, 2587.
- [53] R. D. Septianto, L. Liu, F. Iskandar, N. Matsushita, Y. Iwasa, S. Z. Bisri, *NPG Asia Mater* **2020**, 12, 33.
- [54] H. Kim, M. I. Nugraha, X. Guan, Z. Wang, M. K. Hota, X. Xu, T. Wu, D. Baran, T. D. Anthopoulos, H. N. Alshareef, *ACS Nano* **2021**, 15, 5221.

- [55] M. I. Nugraha, E. Yarali, Y. Firdaus, Y. Lin, A. El-Labban, M. Gedda, E. Lidorikis, E. Yengel, H. Faber, T. D. Anthopoulos, *ACS Appl. Mater. Interfaces* **2020**, 12, 31591.
- [56] D. M. Balazs, B. M. Matysiak, J. Momand, A. G. Shulga, M. Ibáñez, M. V. Kovalenko, B. J. Kooi, M. A. Loi, *Adv. Mater.* **2018**, 30, 1802265.
- [57] D. Shin, Y. Park, H. Jeong, H.-C. V. Tran, E. Jang, S. Jeong, *Adv. Energy Mater.* **2025**, 15, 2304550.
- [58] J. Yang, D. Hahm, K. Kim, S. Rhee, M. Lee, S. Kim, J. H. Chang, H. W. Park, J. Lim, M. Lee, H. Kim, J. Bang, H. Ahn, J. H. Cho, J. Kwak, B. Kim, C. Lee, W. K. Bae, M. S. Kang, *Nat. Commun.* **2020**, 11, 2874.
- [59] H. Jo, M. S. Kang, *Korean J. Chem. Eng.* **2024**, 41, 3431.
- [60] J.-H. Choi, S. W. Lee, J. P. Kar, S. N. Das, J. Jeon, K.-J. Moon, T. I. Lee, U. Jeong, J.-M. Myoung, *J. Mater. Chem.* **2010**, 20, 7393.
- [61] J.-H. Choi, A. T. Fafarman, S. J. Oh, D.-K. Ko, D. K. Kim, B. T. Diroll, S. Muramoto, J. G. Gillen, C. B. Murray, C. R. Kagan, *Nano Lett.* **2012**, 12, 2631.
- [62] M. Zabeli, N. Caka, M. Limani, Q. Kabashi, *WSEAS Trans. Syst. Control* **2008**, 3, 259.
- [63] A. Ali, S. Oh, W. Kim, S. J. Oh, *Korean J. Chem. Eng.* **2024**, 41, 3545.
- [64] H.-Y. Wu, C.-Y. Yang, Q. Li, N. B. Kolhe, X. Strakosas, M.-A. Stoeckel, Z. Wu, W. Jin, M. Savvakis, R. Kroon, D. Tu, H. Y. Woo, M. Berggren, S. A. Jenekhe, S. Fabiano, *Adv. Mater.* **2022**, 34, 2106235.
- [65] A. C. Lazanas, M. I. Prodromidis, *ACS Meas. Sci. Au* **2023**, 3, 162.
- [66] H. Heo, M. H. Lee, J. Yang, H. S. Wee, J. Lim, D. Hahm, J. W. Yu, W. K. Bae, W. B. Lee, M. S. Kang, K. Char, *Nano Lett.* **2017**, 17, 2433.
- [67] J. Koo, J. Yang, B. Cho, H. Jo, K. H. Lee, M. S. Kang, *ACS Appl. Mater. Interfaces* **2018**, 10, 9563.
- [68] W. Alquraishi, Y. Fu, W. Qiu, J. Wang, Y. Chen, L.-A. Kong, J. Sun, Y. Gao, *Org. Electron.* **2019**, 71, 72.
- [69] L.-a. Kong, J. Sun, C. Qian, G. Gou, Y. He, J. Yang, Y. Gao, *Org. Electron.* **2016**, 39, 64.
- [70] D. Lee, K. G. Cho, K. H. Seol, S. Lee, S.-H. Choi, K. H. Lee, *Org. Electron.* **2019**, 71, 266.
- [71] J.-H. Choi, Y. Gu, K. Hong, W. Xie, C. D. Frisbie, T. P. Lodge, *ACS Appl. Mater. Interfaces* **2014**, 6, 19275.
- [72] K. Hong, S. H. Kim, A. Mahajan, C. D. Frisbie, *ACS Appl. Mater. Interfaces* **2014**, 6, 18704.
- [73] J. Yang, M. Lee, S. Y. Park, M. Park, J. Kim, N. Sitapure, D. Hahm, S. Rhee, D. Lee, H. Jo, Y. H. Jo, J. Lim, J. Kim, T. J. Shin, D. C. Lee, K. Kwak, J. S. Kwon, B. Kim, W. K. Bae, M. S. Kang, *Adv. Mater.* **2022**, 34, 2205504.
- [74] H. Ban, Y. Choi, H. G. Lee, W. H. Jung, J. Lim, S.-Y. Cho, *ACS Appl. Nano Mater.* **2024**, 7, 11274.
- [75] J. H. Song, H. Choi, H. T. Pham, S. Jeong, *Nat. Commun.* **2018**, 9, 4267.