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Doped interlayers enabling high-mobility p-type organic transistors with copper contact electrodes[†]

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abstract

Thin-film single crystals of organic semiconductors (OSCs) enable simple and low-cost fabrication of high-mobility organic field-effect transistors (OFETs) via solution processing. However, injection barriers in OFETs limit material selection and device performance. Researchers typically use high-work-function noble metals as contact electrodes for p-type OFETs owing to transport levels well below -5.0 eV for ambient stable OSCs. This raises questions regarding the economic and environmental advantages of OSC-based printed electronics. This study demonstrates high-mobility OFETs with copper contact electrodes using doped interlayers. A poly[bis(4-phenyl)(2,4,6-trimethylphenyl)amine] (PTAA) interlayer was laminated on the OSC single crystals. The PTAA interlayer was chemically doped using aqueous doping solutions, followed by evaporation of copper contact electrodes. Our OFET showed a proper p-type operation with a mobility of $5.0 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ and on-off ratio of approximately 10^4 . This is in contrast to the case without the doped interlayer showing poor performance and a low mobility of $0.2 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$. This study provides new opportunities for designing devices with a high performance, low cost, and material sustainability.

Introduction

Advancements in organic field-effect transistors (OFETs)¹ have opened avenues for various electronic applications, including high-speed integrated circuits^{2–4}, memory devices^{5–7}, and sensors^{8,9}. The use of thin-film single crystals of organic semiconductors (OSCs) enhances the mobility and reliability of OFETs,

thereby providing a way to fabricate integrated devices^{3,10}. High mobilities exceeding $10 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ result from innovations in materials^{11–14} and thin-film fabrication techniques^{10,15–19} for OSCs. These single crystals are formed through a one-shot solution process under ambient conditions, underscoring the environmental and economic sustainability of OSCs.

However, carrier injection into OSCs limits device performance and influences material selection²⁰, which needs to be addressed to expand application areas. OSCs for p-type channels must have the highest occupied molecular orbital (HOMO) energy levels well below -5.0 eV vs. vacuum to ensure ambient stability²¹. This requirement explains the reliance on high-work-function noble metals such as platinum and gold as electrodes for proper transistor operation, especially in high-mobility OFETs, which poses challenges for economic and material sustainability²². To use an abundant metal, such as copper, with a work function (WF) of 4.6 eV, careful alignment of energy levels at the contacts must be addressed^{23,24}.

Engineering at the electrode-semiconductor interface is crucial for improving carrier injection into OSCs. Self-assembled monolayers modify the electrode WF, reducing contact resistance in bottom-contact geometries². Although this method was effective for copper electrodes^{24,25}, the reported contact resistance was approximately $1 \text{ M}\Omega\text{cm}$, far exceeding the optimal range necessary for high-mobility OFETs. An alternative approach involves the use of multiple layers of OSCs with varying energy levels to reduce injection barriers²⁶. This strategy of employing interlayers to facilitate charge carrier injection and extraction is, in fact, a well-established and highly effective technique in the field of photovoltaics, particularly for high-performance organic solar cells^{27–30}. In p-type transistors, molecules with large electron affinities have been introduced between the electrodes and active layers to create contact-doping effects³¹. However, the applicability of this strategy to copper electrodes with p-type OSCs remains uncertain owing to the large difference in energy levels. Previous reports were mostly limited to OSCs with a shallow HOMO

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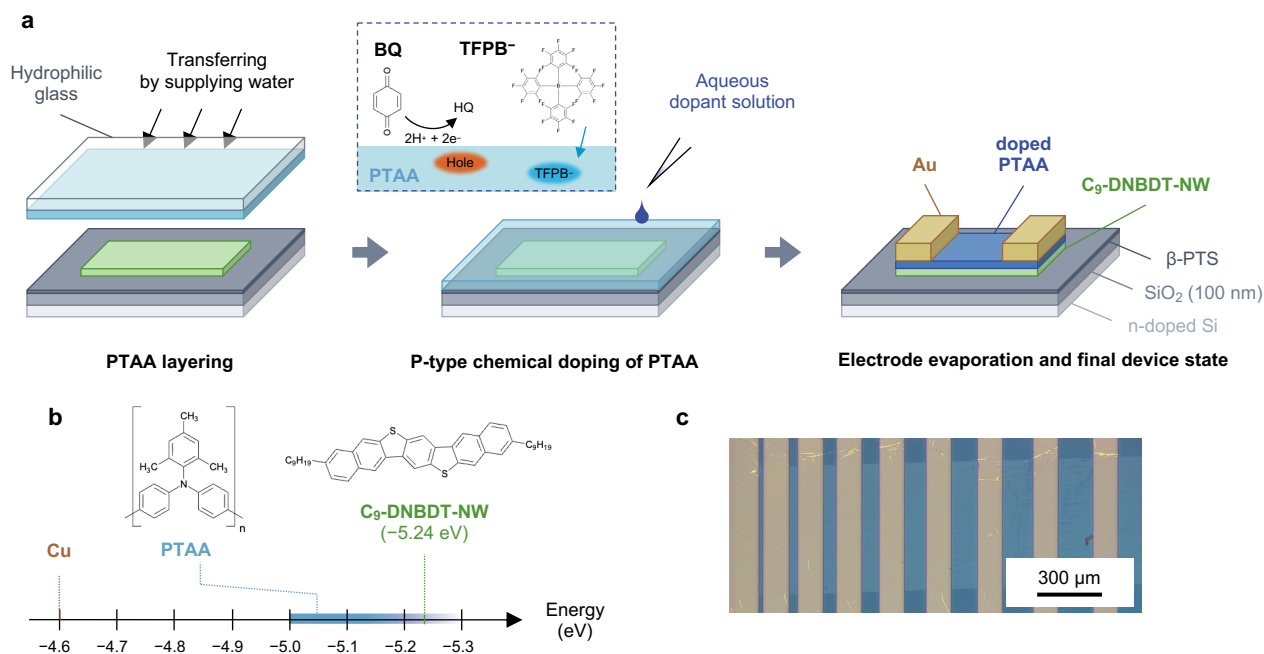


Fig. 1 (a) Schematics of the device fabrication, illustrating the process of transfer of poly[bis(4-phenyl)(2,4,6-trimethylphenyl)amine] (PTAA) layer on C₉-DNBDT-NW single crystals, chemical doping, and electrode deposition. (b) Chemical structures of PTAA and C₉-DNBDT-NW together with the transport energy levels of employed materials. (c) A polarized microscopy image of the fabricated single-crystal organic field-effect transistors.

and instability in air^{32,33}. For bottom-contact geometries, treatment of copper electrodes with molecular acceptors increases the work function of the electrodes and the mobility of OFETs operating under ambient conditions³⁴. However, it is unclear whether the contact resistance can go below 1 MΩcm with this method. Recent advances in efficient chemical doping and highly doped materials^{35–39} may provide a way to dope semiconductor active layers and thus decrease the injection barriers. However, this approach can cause undesirable doping of channels and a decrease in the on-off ratio. Thus, the use of copper or low-work-function metals as contact electrodes in high-performance p-type OFETs remains challenging.

Here, we propose a new contact structure for OFETs that enables high-mobility operation using copper electrodes by introducing a doped polymer interlayer. For the 3,11-dinonyldinaphtho[2,3-*d*:2',3'-*d'*]benzo[1,2-*b*:4,5-*b'*]dithiophene(C₉-DNBDT-NW)¹⁰ single-crystal active layer, we selected the polymeric semiconductor poly[bis(4-phenyl)(2,4,6-trimethylphenyl)amine] (poly(triarylamine) (PTAA)) as the interlayer material. This choice was motivated by two key factors. First, PTAA's deep HOMO level of approximately -5 eV is well-aligned with that of C₉-DNBDT-NW, which is expected to reduce the hole injection barrier from the electrode. Second, PTAA itself possesses a field-effect mobility of 0.003 cm² V⁻¹ s⁻¹⁴⁰, which is low relative to C₉-DNBDT-NW, ensuring that its insertion does not interfere with FET on/off operation. In our structure, a thin PTAA interlayer was stacked on the C₉-DNBDT-NW single crystal. After the chemical doping of the PTAA interlayer using an aqueous solution³⁹, copper contact electrodes were thermally deposited. Our OFET showed

proper p-type operation with a mobility of 5.0 cm² V⁻¹ s⁻¹ and an on-off ratio of approximately 10⁴, which is dramatically improved compared to the case without the doped interlayer. Here, the moderately shallow doping level of the PTAA interlayer, as revealed by X-ray photoelectron spectroscopy (XPS) and ultraviolet photoelectron spectroscopy (UPS), enabled the proper normally-off operation. The balanced on-off ratios and mobility suggest that the rational use of doped polymer interlayers enables the employment of abundant metals, such as copper, in OFET contact electrodes.

Results and Discussions

OFET Fabrication and Characterization

To investigate the effect of the PTAA interlayer between the source/drain electrodes and C₉-DNBDT-NW active layer, we fabricated single-crystal OFETs, as illustrated in Fig. 1. A thin-film single crystal of C₉-DNBDT-NW was deposited via the continuous edge-casting method^{10,15} (see Methods section for details). The schematic of the fabrication process is presented in Fig. 1a. The PTAA interlayer was spin-coated onto a hydrophilic glass substrate with a thickness of less than 10 nm (Fig. S1) and subsequently transferred onto the C₉-DNBDT-NW layer. In this process, the PTAA interlayer was peeled off the hydrophilic glass substrate by supplying water⁴¹. The PTAA interlayer was then chemically doped by treatment with an aqueous solution of benzoquinone (BQ) and tetrakis(pentafluorophenyl)borate anion (TFPB⁻). The doping process involves redox reactions between BQ and the organic semiconductor, introducing holes into the material. This is accompanied by the incorporation of

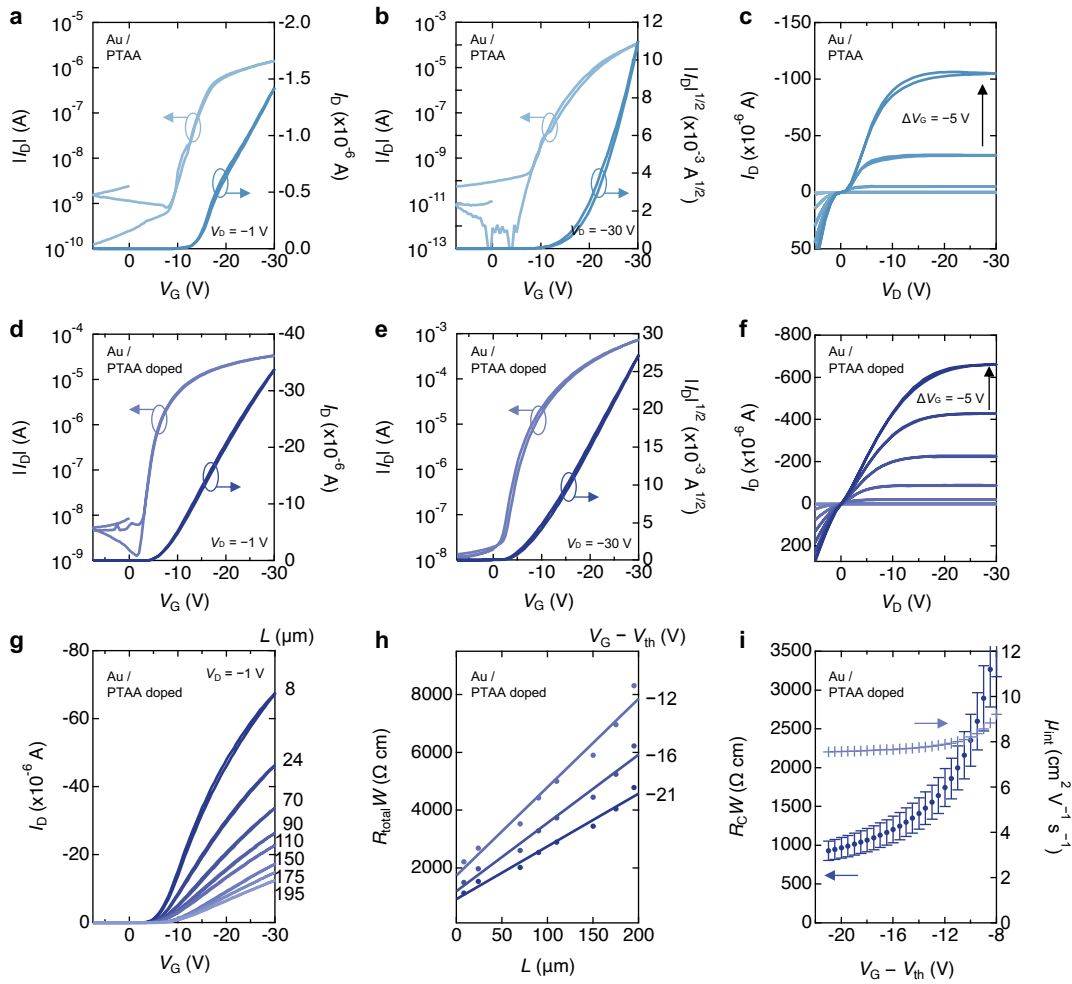


Fig. 2 Comparison of OFET characteristics using Au electrodes and PTAA interlayers with and without chemical doping. Transfer curves in the (a) linear regime (V_D at -1 V) and (b) saturation regime (V_D at -30 V) and (c) the output characteristics of the OFET fabricated without chemical doping. The channel length (L) is $65\ \mu\text{m}$. Transfer curves in the (d) linear regime (V_D at -1 V) and (e) saturation regime (V_D at -30 V) and (f) the output characteristics of the OFET fabricated using the chemical doping process. L is $70\ \mu\text{m}$. In (c) and (f), employed V_G values are -30 V, -25 V, -20 V, -15 V, -10 V, -5 V, 0 V, and 5 V. (g) Transfer curves in the linear regime with different L for OFET with doped PTAA and (h) corresponding transfer length method (TLM) plots at various $V_G - V_{th}$. (i) Dependence of $R_C W$ and μ_{int} on $V_G - V_{th}$.

TFPB⁻ anions for charge compensation. Finally, the source/drain electrodes were fabricated via thermal deposition using a shadow mask. All devices had a channel width (W) of $600\ \mu\text{m}$, while the channel length L was varied.

The effect of chemical doping was assessed using OFETs with gold source/drain electrodes. Fig. 2a-c show representative characteristics of our OFET with an undoped PTAA interlayer. The mobilities extracted from transfer characteristics were $\mu_{lin} = 0.23\ \text{cm}^2\ \text{V}^{-1}\ \text{s}^{-1}$ and $\mu_{sat} = 5.1\ \text{cm}^2\ \text{V}^{-1}\ \text{s}^{-1}$ in the linear and saturation regimes, respectively. In the linear regime, a large kink was observed in the current level as the gate voltage (V_G) approached the threshold voltage (V_{th})⁴². The output characteristics exhibited S-shaped curves at low drain voltages (V_D), indicating high contact resistances. Fig. 2d-f show the characteristics of an OFET with a doped PTAA interlayer. Compared to the undoped case, mobilities were increased, reaching μ_{lin} of $4.8\ \text{cm}^2\ \text{V}^{-1}\ \text{s}^{-1}$ and μ_{sat} of $9.2\ \text{cm}^2\ \text{V}^{-1}\ \text{s}^{-1}$. We applied the

transfer length method (TLM) to evaluate the contact resistance (R_C) using doped OFETs with channel lengths of 8, 24, 70, 90, 110, 150, 175, and $190\ \mu\text{m}$ (Fig. 2g). Fig. 2h shows the width-normalized total resistance ($R_{total} \cdot W$) as a function of L for various $V_G - V_{th}$ values. We estimated $R_C \cdot W$ and intrinsic mobility (μ_{int}) from the intercept and slope of this plot, respectively, and plotted it against $V_G - V_{th}$ in Fig. 2i. At the highest carrier density ($V_G - V_{th} = -21$ V), $R_C \cdot W$ was estimated to be $930 \pm 130\ \Omega\text{cm}$. These results suggest that doping the PTAA interlayer effectively reduces contact resistance in OFETs, enabling sufficiently low R_C values for high-mobility operation. While this value is not particularly remarkable compared to conventional single-crystal OFETs with Au contact electrodes (typically less than $\times 10^3\ \Omega\text{cm}$)⁴³, it is noteworthy in the context of Cu contact electrodes, as discussed in the following sections.

We observed enhanced injection properties using doped interlayers even when copper contact electrodes were employed.

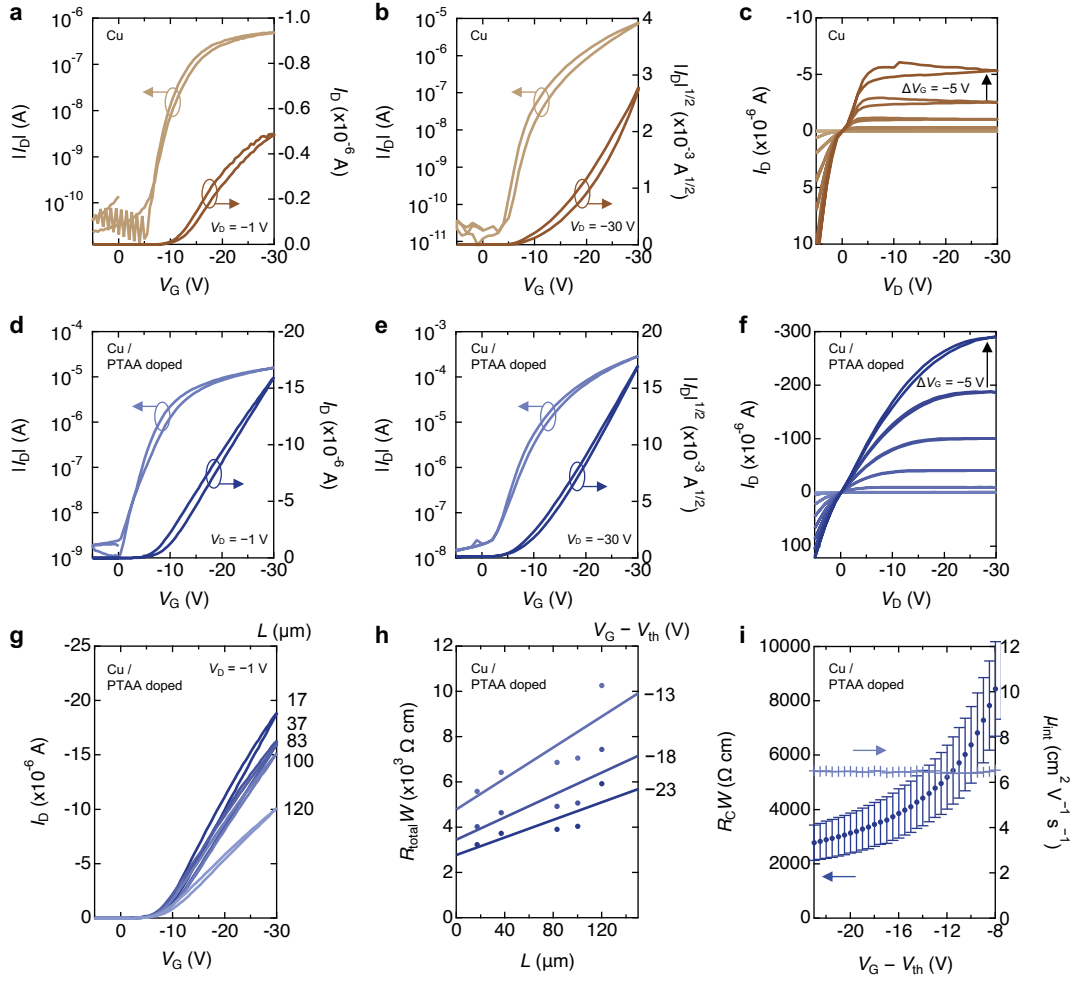


Fig. 3 Comparison of OFET characteristics using Cu electrodes with and without doped PTAA interlayer. Transfer curves in the (a) linear regime (V_D at -1 V) and (b) saturation regime (V_D at -30 V) and (c) the output characteristics of the OFET without PTAA interlayer. L is 84 μm . Transfer curves in the (d) linear regime (V_D at -1 V) and (e) saturation regime (V_D at -30 V) and (f) the output characteristics of the OFET with doped PTAA interlayer. L is 83 μm . In (c) and (f), the employed V_G values are -30 V, -25 V, -20 V, -15 V, -10 V, -5 V, 0 V, and 5 V. (g) Transfer curves in the linear regime with different L in OFETs with the doped PTAA interlayer and (h) corresponding TLM plots at various $V_G - V_{th}$. (i) Dependence of $R_C W$ and μ_{int} on $V_G - V_{th}$.

Typically, copper electrodes have a WF of 4.6 eV^{24} , which is inadequate for effective hole injection into p-type OSCs with ambient stability. Indeed, poor performance was observed in our OFET when copper was directly deposited on C₉-DNBDT-NW, as shown in Fig 3a-c. In this case, μ_{lin} was $9.4 \times 10^{-2} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ and μ_{sat} was $2.0 \times 10^{-1} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, which are lower than the intrinsic mobility of C₉-DNBDT-NW. In the linear regime, a distinct kink was observed in the transfer curve. The output characteristics exhibited current saturation at low V_D . These behaviors suggest that transistor performance suffers significantly from the large injection barrier between the copper electrodes and C₉-DNBDT-NW layer. Assuming that contact limits the device performance in the linear regime, $R_C \cdot W$ is approximately $1.2 \times 10^5 \Omega \text{ cm}$. The device performance was significantly improved by stacking a doped PTAA interlayer between the copper electrode and C₉-DNBDT-NW layer, as shown in Fig. 3 d-f. In this configuration, we observed increases in μ_{lin} to $2.8 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ and μ_{sat} to $5.0 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$. Current saturation in the output

characteristics occurred when V_D and V_G approached each other, which is in agreement with the standard transistor operation. The on-off ratio of our device remained high at approximately 10^4 . Here, the PTAA layer contributes minimally to the off currents, especially given the substantial mobility difference between PTAA ($3 \times 10^{-3} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$)⁴⁰ and C₉-DNBDT-NW. While too high doping levels of the C₉-DNBDT-NW channel would increase the off currents, our doping process resulted in sufficiently low doping levels to maintain the on-off ratio, as discussed in detail later. We applied the TLM method to evaluate the contact resistance of copper-based OFETs with the doped PTAA interlayer, similar to our assessment of gold electrodes. Fig. 3g and h show the transfer curve in the linear region of the OFETs with varying channel lengths and the relationship between $R_{total} \cdot W$ and L at various values of $V_G - V_{th}$. The dependences of $R_C \cdot W$ and μ_{int} on $V_G - V_{th}$ are shown in Fig. 3i. At the maximum carrier density ($V_G - V_{th} = -23$ V), $R_C \cdot W$ was estimated to be $(2.8 \pm 0.7) \times 10^3 \Omega \text{ cm}$, which is, to the best of our knowledge, the lowest value

reported for copper-based OFETs operating in air. These findings demonstrate that our doped interlayer substantially enhances the carrier injection properties, even when the electrodes have relatively low WFs.

Photoelectron measurements of the doped PTAA layer

To clarify the structure and role of the PTAA layer, XPS and UPS measurements were performed. Fig. 4a shows the XPS survey spectra of PTAA films. The pristine PTAA spectrum reveals peaks for C 1s, N 1s, Au 4f, and O 1s. In contrast, the spectrum of PTAA doped using an aqueous solution of BQ and Li-TFPB shows a new F 1s peak, confirming the incorporation of TFPB[−] into the PTAA film. Table 1 lists the relative quantitative analysis results for each element detected from the narrow spectra (Fig. S2). The ratios of N and F indicate that TFPB[−] is present at approximately one TFPB[−] for every five PTAA monomers around the surface of the thin film based on the molecular structures of the anion and PTAA.

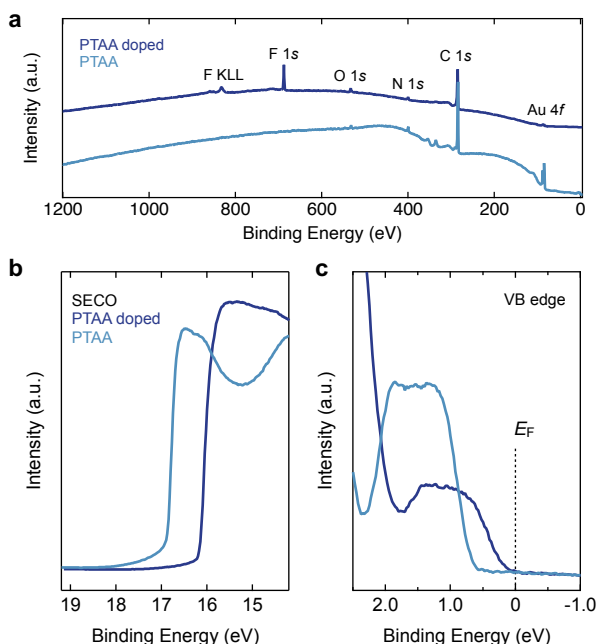


Fig. 4 (a) X-ray photoelectron spectroscopy survey spectrum of thin films of undoped and doped PTAA. KLL shows peaks originating from KLL Auger transitions. UPS spectra of the region of (b) secondary electron cutoff (SECO) and (c) the edge of HOMO density of states for undoped and doped PTAA thin films. (The binding energy of the Fermi level is aligned to 0 eV.)

Table 1 Relative atomic concentrations (%) of elements detected in the photoelectron escape depth range in PTAA thin films before and after doping.

sample	C	N	F	Au	O
PTAA (pristine)	69.4	3.9	-	2.1	1.4
PTAA (doped)	80.1	3.4	13.7	0.2	2.6

Fig. 4b and c show the UPS spectra of undoped and doped PTAA thin films. The origin of the binding energy aligns with the

Fermi energy (E_F) as determined by the Fermi edge observed in a gold electrode. Based on the position of secondary electron cut-off (SECO), WF of PTAA thin films were determined to be 4.3 eV and 5.0 eV, respectively. The increased WF after the treatment with solutions containing BQ and Li-TFPB indicates effective doping, which is consistent with the reported oxidation capability of BQ³⁹. Notably, after the doping process, the edge of the HOMO density of states was close to E_F .

Considering that semiconductor doping levels significantly influence injection barriers at metal/semiconductor interfaces, our doping process reduces the barrier height between the copper electrodes and PTAA layers. Although high doping levels can lower this barrier height, excessive doping in the PTAA layer in contact with the C₉-DNBDT-NW active layer could hinder normally-off operation. Here, a WF of 5.0 eV in the PTAA layer may not result in the excessive doping of C₉-DNBDT-NW with an ionization potential of 5.24 eV¹⁴. Thus, our doping process achieves a balance between injection properties from copper electrodes and the normally-off operation of our OFETs. In Fig. 3b, the current in the subthreshold region begins to rise from around V_G of 0 V, supporting the conclusion that the Fermi energy is close to optimal. This interpretation is consistent with previous reports showing that the onset voltage shifts with doping level in similar configurations⁴⁴. Adjusting the WF is necessary depending on the interlayer and active layer choice. Using aqueous solutions for chemical doping simplifies WF control through pH adjustments³⁹. Considering that doped PTAA has been used as a hole-transporting layer in perovskite solar cells⁴⁵, this strategy may be applicable to a broad range of p-type semiconductors and related devices.

Conclusion

This paper presents a novel approach for fabricating high-performance electronic devices using extended choice for electrode materials. The PTAA interlayer was stacked between copper electrodes and thin-film single crystals of C₉-DNBDT-NW, which was chemically doped using aqueous doping solutions. The employment of doped interlayer enhanced hole injection properties and mobility in p-type OFETs. A contact resistance of $2.8 \times 10^3 \Omega \text{cm}$ and μ_{sat} of $5.0 \text{ cm}^2 \text{V}^{-1} \text{s}^{-1}$ were observed, indicating significant improvements over conventional devices using copper electrodes. The shallow yet effective chemical doping successfully reduces the injection barrier from the copper electrodes while maintaining high on-off ratios. These findings can advance OSC-based devices by expanding material selection, thus enhancing cost efficiency and environmental sustainability.

Methods

SC-OSC Fabrication

An n-type doped silicon substrate with a 100 nm SiO₂ layer was treated with a self-assembled monolayer of 2-(phenylhexyl)trimethoxysilane. SCs of C₉-DNBDT-NW were fabricated from a 0.02 wt% 3-chlorothiophene solution using the continuous edge-casting method¹⁵. During this process, the substrate temperature was precisely controlled to obtain the SC domain within the monolayer.

Interlayer Fabrication

An EAGLE XG glass substrate (Corning Inc.) was treated with UV/O₃ for 15 min to enhance its hydrophilicity. The polymer interlayers, poly[bis(4-phenyl)(2,4,6-trimethylphenyl)amine](PTAA) (Solaris Chem Inc.), were dissolved in toluene at a concentration of 0.2 wt% and spin-coated onto the UV/O₃ treated glass substrates. These PTAA thin films were placed directly above the C₉-DNBDT-NW layers. The PTAA interlayers were fabricated by peeling them off the hydrophilic glass substrate using deionized water followed by annealing at 80 °C under vacuum to remove the solvent.

Interlayer Doping and SC-OFET Fabrication

To dope the interlayers, we immersed them in aqueous dopant solutions under ambient conditions. The doping solutions were prepared by dissolving 10 mM BQ and 10 mM lithium TFPB in a pH= 2.0 aqueous solution. After immersion, the interlayers were washed with deionized water, and the residual solvent was removed using an N₂ flow.

Au was deposited using a metal mask to create the source/drain electrodes. C₉-DNBDT-NW and PTAA layers were patterned via dry-etching with a yttrium-aluminum-garnet laser (266 nm).

Electrical Measurement

Electrical measurements were performed using a semiconductor parameter analyzer (Keithley 4200-SCS) in the dark under ambient conditions. The mobilities in the linear (μ_{lin}) and saturation (μ_{sat}) regimes were extracted from the transfer characteristics using the following equations:

$$I_{\text{D,lin}} = \mu_{\text{lin}} \frac{WC_i}{L} (V_{\text{G}} - V_{\text{th}}) V_{\text{D}}$$

$$I_{\text{D,sat}} = \mu_{\text{sat}} \frac{WC_i}{2L} (V_{\text{G}} - V_{\text{th}})^2$$

where I_{D} , L , W , C_i , V_{G} , V_{th} , and V_{D} are the drain current, channel length, channel width, capacitance per unit area, gate voltage, threshold voltage, and drain voltage, respectively. $C_i = 34.5 \text{ nFcm}^{-2}$ was used for all devices.

XPS and UPS Analysis

XPS and UPS experiments were performed using a KRATOS ULTRA 2 instrument. XPS employed monochromatic Al K α X-rays for a wide scan from 0 to 1200 eV. For UPS, we utilized photons at 21.2 eV from a He discharge lamp. The binding energies in the UPS experiments were calibrated using the Fermi edge of the metal substrates. A bias voltage of -8.0 V was applied to the sample during UPS measurements.

Author Contributions

A.M. fabricated and evaluated the transistors. Y.Y. contributed to the conceptualization of this work. T. S. contributed to the evaluation of the transistors. M.I. contributed to the development of the doping process. S.W. and J.T. supervised the study. A. M. and Y. Y. wrote the manuscript.

Conflicts of interest

The authors declare no conflict of interest.

Data availability

The data supporting this work are available in the main text and Supplementary Information.

Acknowledgments

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Notes and references

- 1 A. Nawaz, L. Mercas, L. M. Ferro, P. Sonar and C. C. Bufon, *Advanced Materials*, 2023, **35**, 2204804.
- 2 J. W. Borchert, U. Zschieschang, F. Letzkus, M. Giorgio, R. T. Weitz, M. Caironi, J. N. Burghartz, S. Ludwigs and H. Klauk, *Science Advances*, 2020, **6**, eaaz5156.
- 3 X. Wei, S. Kumagai, T. Makita, K. Tsuzuku, A. Yamamura, M. Sasaki, S. Watanabe and J. Takeya, *Communications Materials*, 2023, **4**, 4.
- 4 F. Parenti, R. Sargeni, E. Dimaggio, F. Pieri, F. Fabbri, T. Losi, F. A. Viola, A. Bala, Z. Wang, A. Kis *et al.*, *Nano Letters*, 2024, **24**, 15870–15877.
- 5 J. Aimi, C.-T. Lo, H.-C. Wu, C.-F. Huang, T. Nakanishi, M. Takeuchi and W.-C. Chen, *Advanced Electronic Materials*, 2016, **2**, 1500300.
- 6 Y. Ni, Y. Wang and W. Xu, *Small*, 2021, **17**, 1905332.
- 7 K.-J. Baeg, D. Khim, J. Kim, B.-D. Yang, M. Kang, S.-W. Jung, I.-K. You, D.-Y. Kim and Y.-Y. Noh, *Advanced Functional Materials*, 2012, **22**, 2915–2926.
- 8 C. Zhang, P. Chen and W. Hu, *Chemical Society Reviews*, 2015, **44**, 2087–2107.
- 9 S. Yuvaraja, A. Nawaz, Q. Liu, D. Dubal, S. G. Surya, K. N. Salama and P. Sonar, *Chemical Society Reviews*, 2020, **49**, 3423–3460.
- 10 S. Kumagai, A. Yamamura, T. Makita, J. Tsurumi, Y. Y. Lim, T. Wakimoto, N. Isahaya, H. Nozawa, K. Sato, M. Mitani, T. Okamoto, S. Watanabe and J. Takeya, *Scientific Reports*, 2019, **9**, 1–8.
- 11 K. Takimiya, H. Ebata, K. Sakamoto, T. Izawa, T. Otsubo and Y. Kunugi, *Journal of the American Chemical Society*, 2006, **128**, 12604–12605.
- 12 H. Ebata, T. Izawa, E. Miyazaki, K. Takimiya, M. Ikeda, H. Kuwabara and T. Yui, *Journal of the American Chemical Society*, 2007, **129**, 15732–15733.
- 13 T. Yamamoto and K. Takimiya, *Journal of the American Chemical Society*, 2007, **129**, 2224–2225.
- 14 C. Mitsui, T. Okamoto, M. Yamagishi, J. Tsurumi, K. Yoshimoto, K. Nakahara, J. Soeda, Y. Hirose, H. Sato, A. Yamano, T. Uemura and J. Takeya, *Advanced Materials*, 2014, **26**, 4546–4551.
- 15 J. Soeda, T. Uemura, T. Okamoto, C. Mitsui, M. Yamagishi and J. Takeya, *Applied Physics Express*, 2013, **6**, 076503.
- 16 B. Peng, Z. Wang and P. K. L. Chan, *Journal of Materials Chemistry C*, 2016, **4**, 8628–8633.
- 17 Z. Zhang, B. Peng, X. Ji, K. Pei and P. K. L. Chan, *Advanced*

- Functional Materials*, 2017, **27**, 1703443.
- 18 A. Yamamura, S. Watanabe, M. Uno, M. Mitani, C. Mitsui, J. Tsurumi, N. Isahaya, Y. Kanaoka, T. Okamoto and J. Takeya, *Science Advances*, 2018, **4**, year.
 - 19 M. Chen, B. Peng, S. Huang and P. K. L. Chan, *Advanced Functional Materials*, 2020, **30**, 1905963.
 - 20 J. W. Borchert, R. T. Weitz, S. Ludwigs and H. Klauk, *Advanced Materials*, 2022, **34**, 2104075.
 - 21 P. A. Bobbert, A. Sharma, S. G. Mathijssen, M. Kemerink and D. M. de Leeuw, *Advanced Materials*, 2012, **24**, 1146–1158.
 - 22 F. Torricelli, I. Alessandri, E. Macchia, I. Vassalini, M. Madaloni and L. Torsi, *Advanced Materials Technologies*, 2022, **7**, 2100445.
 - 23 C.-a. Di, G. Yu, Y. Liu, X. Xu, D. Wei, Y. Song, Y. Sun, Y. Wang, D. Zhu, J. Liu *et al.*, *Journal of the American Chemical Society*, 2006, **128**, 16418–16419.
 - 24 C.-H. Kim, H. Hlaing, J.-A. Hong, J.-H. Kim, Y. Park, M. M. Payne, J. E. Anthony, Y. Bonnassieux, G. Horowitz and I. Kyymissis, *Advanced Materials Interfaces*, 2015, **2**, 1400384.
 - 25 K. Patrikar, U. Bothra, V. R. Rao and D. Kabra, *Advanced Materials Interfaces*, 2022, **9**, 2101377.
 - 26 N. B. Kotadiya, H. Lu, A. Mondal, Y. Ie, D. Andrienko, P. W. Blom and G.-J. A. Wetzelaer, *Nature Materials*, 2018, **17**, 329–334.
 - 27 X. Wang, J. Tian, Z. You, L. Lei, A. Ge and Y. Liu, *Chinese Journal of Chemistry*, 2024, **42**, 2979–2986.
 - 28 Z. You, J. Wen, W. Liu, Z. Fink, X. Wu, H.-G. Seong, Y. Wang, L. Zhang, X. Wang, T. P. Russell *et al.*, *Advanced Materials*, 2025, **37**, 2500450.
 - 29 Z. Chen, Q. Li, H. Tang, J. Wen, Y. Zhong, J. Zhang, K. Han and Y. Liu, *Angewandte Chemie International Edition*, 2025, **64**, e202424502.
 - 30 Z. You, A. Gao and Y. Liu, *Chemical Communications*, 2025.
 - 31 Y. Kim, S. Chung, K. Cho, D. Harkin, W.-T. Hwang, D. Yoo, J.-K. Kim, W. Lee, Y. Song, H. Ahn *et al.*, *Advanced Materials*, 2019, **31**, 1806697.
 - 32 J. Li, X.-W. Zhang, L. Zhang, H. Zhang, X.-Y. Jiang, W.-Q. Zhu, Z.-L. Zhang *et al.*, *Synthetic metals*, 2010, **160**, 376–379.
 - 33 Y. Su, M. Wang, F. Xie, J. Chen, W. Xie, N. Zhao and J. Xu, *Organic Electronics*, 2013, **14**, 775–781.
 - 34 J. Frisch, H. Glowatzki, S. Janietz and N. Koch, *Organic Electronics*, 2009, **10**, 1459–1465.
 - 35 C. G. Tang, M. C. Ang, K.-K. Choo, V. Keerthi, J.-K. Tan, M. N. Syafiqah, T. Kugler, J. H. Burroughes, R.-Q. Png, L.-L. Chua *et al.*, *Nature*, 2016, **539**, 536–540.
 - 36 R. Kroon, D. Kiefer, D. Stegerer, L. Yu, M. Sommer and C. Müller, *Advanced Materials*, 2017, **29**, 1700930.
 - 37 Y. Yamashita, J. Tsurumi, M. Ohno, R. Fujimoto, S. Kumagai, T. Kurosawa, T. Okamoto, J. Takeya and S. Watanabe, *Nature*, 2019, **572**, 634–638.
 - 38 I. E. Jacobs, Y. Lin, Y. Huang, X. Ren, D. Simatos, C. Chen, D. Tjhe, M. Statz, L. Lai, P. A. Finn *et al.*, *Advanced Materials*, 2022, **34**, 2102988.
 - 39 M. Ishii, Y. Yamashita, S. Watanabe, K. Ariga and J. Takeya, *Nature*, 2023, **622**, 285–291.
 - 40 X. Lu, L. Majewski and A. Song, *Organic Electronics*, 2008, **9**, 473–480.
 - 41 T. Makita, S. Kumagai, A. Kumamoto, M. Mitani, J. Tsurumi, R. Hakamatani, M. Sasaki, T. Okamoto, Y. Ikuhara, S. Watanabe *et al.*, *Proceedings of the National Academy of Sciences*, 2020, **117**, 80–85.
 - 42 I. McCulloch, A. Salleo and M. Chabiny, *Science*, 2016, **352**, 1521–1522.
 - 43 A. Yamamura, S. Watanabe, M. Uno, M. Mitani, C. Mitsui, J. Tsurumi, N. Isahaya, Y. Kanaoka, T. Okamoto and J. Takeya, *Science Advances*, 2018, **4**, eaao5758.
 - 44 H. Guo, C.-Y. Yang, X. Zhang, A. Motta, K. Feng, Y. Xia, Y. Shi, Z. Wu, K. Yang, J. Chen *et al.*, *Nature*, 2021, **599**, 67–73.
 - 45 T. Wang, Y. Zhang, W. Kong, L. Qiao, B. Peng, Z. Shen, Q. Han, H. Chen, Z. Yuan, R. Zheng *et al.*, *Science*, 2022, **377**, 1227–1232.