Supplementary Material for

High-k Organic-Inorganic Hybrid Dielectric material for Flexible Thin-Film Transistors and Printed Logic Circuits

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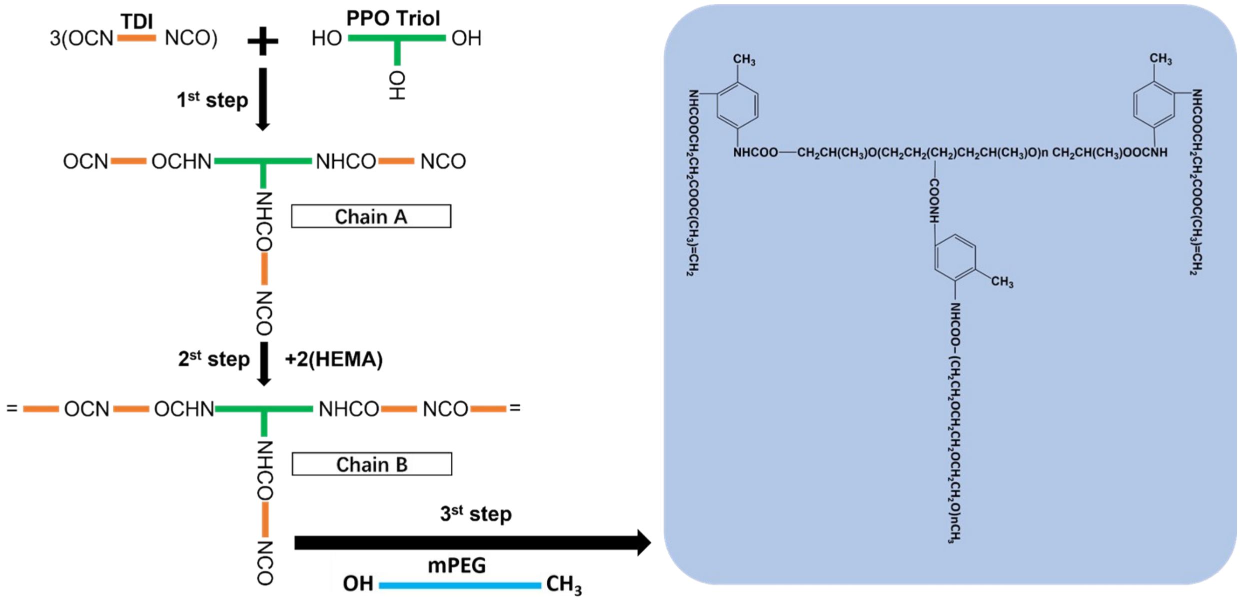


Fig. S1. The chemical structure and synthesis pathway of AUP

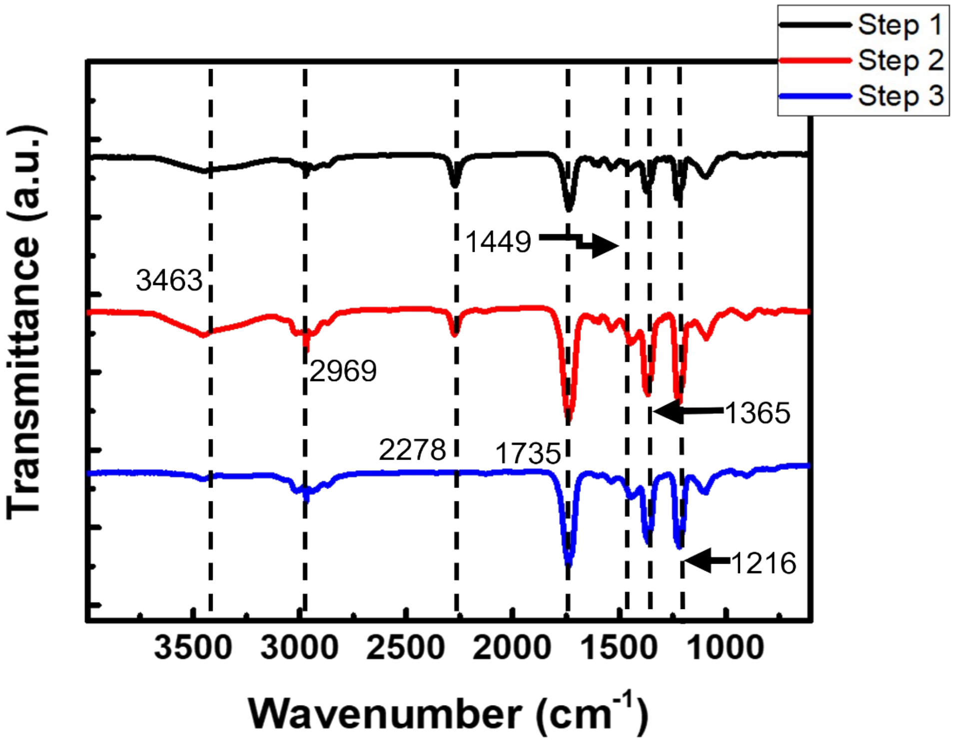
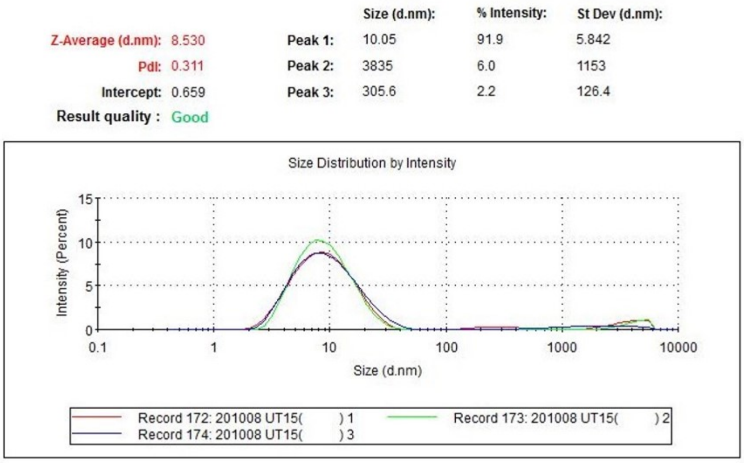


Fig. S2. FTIR spectra of the three steps in the synthesis process of AUP component in O-I hybrid solution.

AUP@SiOx



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                                     Size (d.nm):   % Intensity:   St Dev (d.nm):
Z-Average (d.nm): 8.530    Peak 1:   10.05          91.9           5.842
             PdI: 0.311    Peak 2:   3835           6.0            1153
       Intercept: 0.659    Peak 3:   305.6          2.2            126.4
 Result quality : Good
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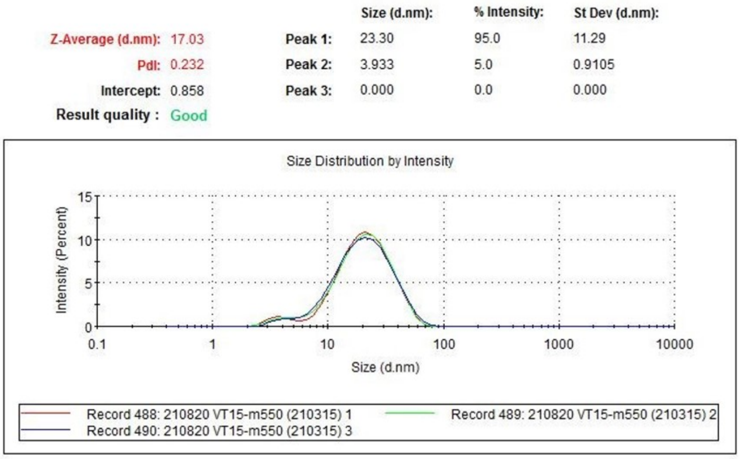
AUP@SiOx-184



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                                     Size (d.nm):   % Intensity:   St Dev (d.nm):
Z-Average (d.nm): 17.03    Peak 1:   23.30          95.0           11.29
             PdI: 0.232    Peak 2:   3.933          5.0            0.9105
       Intercept: 0.858    Peak 3:   0.000          0.0            0.000
 Result quality : Good
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Fig. S3. The DLS about AUP@SiOx and AUP@SiOx-184.

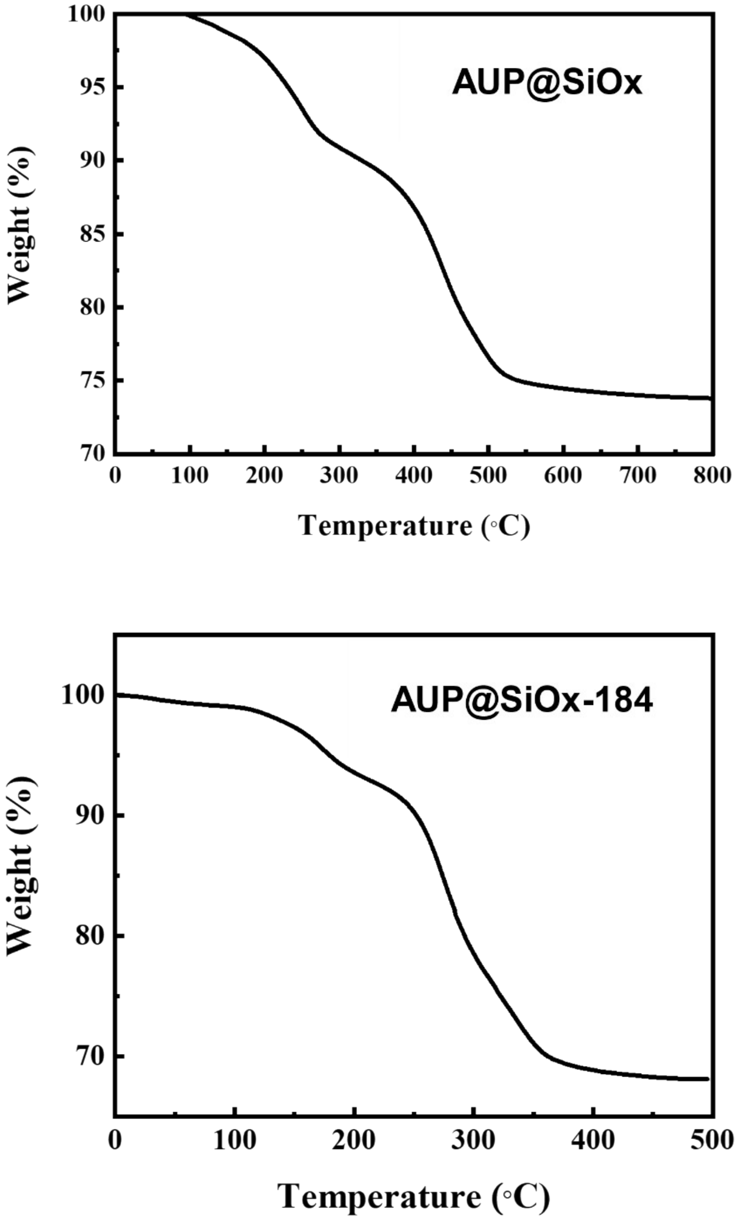


Fig S4. TGA data for the AUP@SiOx and AUP@SiOx-184 solutions.

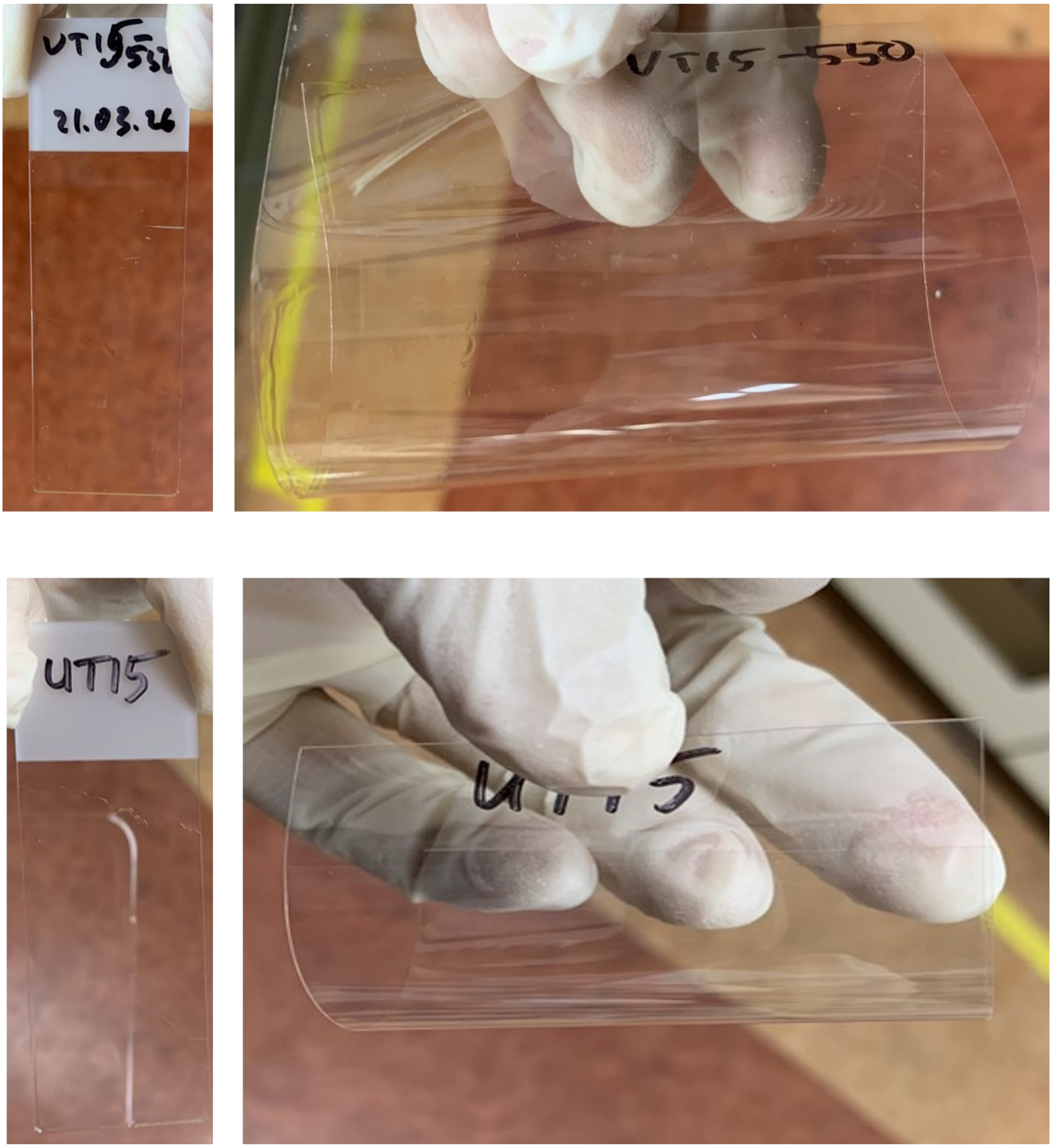


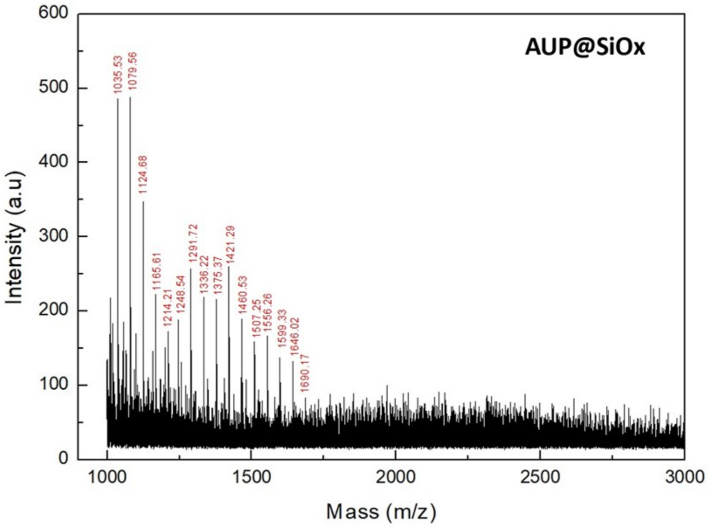
Fig S5. Coating tests of AUP@SiOx-184 and AUP@SiOx solutions on glass and PET substrates, respectively.

Table S1. Hardness testing after coating and curing of AUP@SiOx-184 and AUP@SiOx solutions on glass and PET substrates, respectively.

O-I hybrid coating film	**Pencil hardness on glass slide (H)**	**Pencil hardness on PET film (H)**
AUP@SiOx	7	3
AUP@SiOx-184	7	4

MALDI-TOF-MS analysis of AUP@SiOx O-I hybrid Solutions

- The range of Mw: 1035.53 – 1690.17g/mol
- The average Mw: 1079.56 g/mol



MALDI-TOF-MS analysis of AUP@SiOx-184 O-I hybrid Solutions

- The range of Mw: 942.97 – 2026.14g/mol
- The average Mw: 1492.94 g/mol

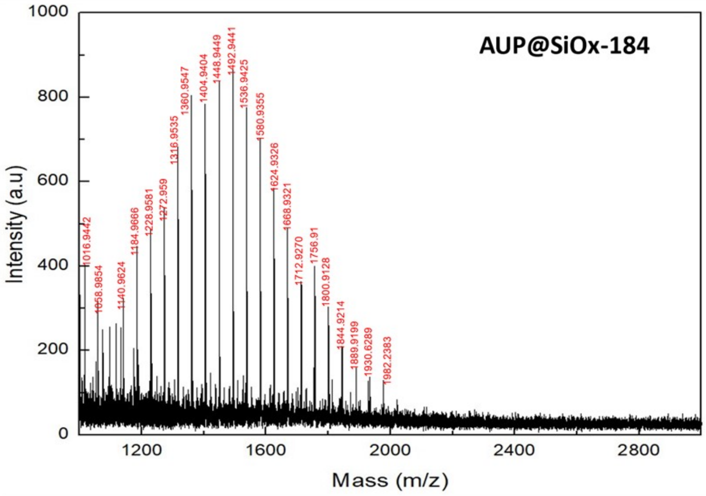
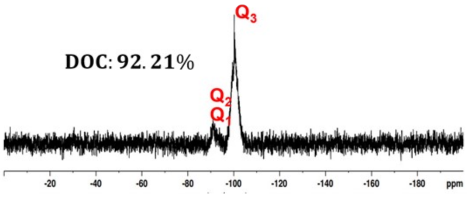


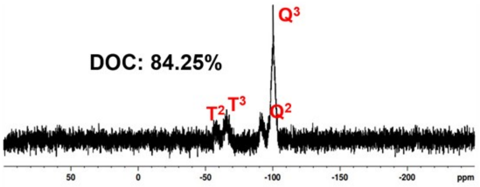
Fig S6. The molecular weight range of AUP@SiOx and AUP@SiOx-184.

^{29}Si – NMR analysis of AUP@SiOx O-I hybrid solution

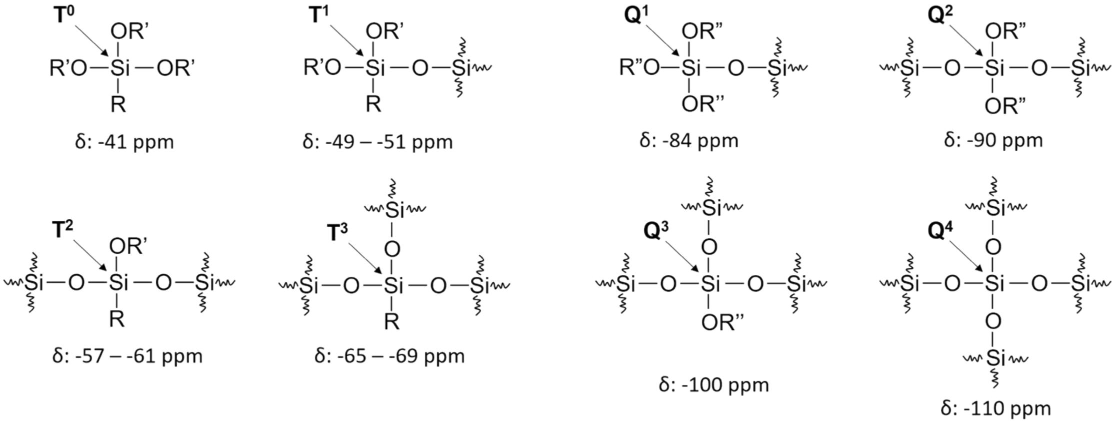


- **Q^m** denote the silicon from tetrafunctional alkoxysilanes .
- **Q^2** indicates two linkages (linear groups).
- **Q^3** indicates three linkages

^{29}Si – NMR analysis of AUP@SiOx-184 O-I hybrid solution



- **T^n** and **Q^m** denote the silicon from trifunctional alkoxysilanes and tetrafunctional alkoxysilanes, respectively.
- **T^2** illustrates the middle groups of chains or cycles.
- **T^3** shows fully branched sites.
- **Q^2** indicates two linkages (linear groups).
- **Q^3** indicates three linkages



$$\mathrm{DOC} = \frac{Q^1 + 2Q^2 + 3Q^3 + T^1 + 2T^2 + 3T^3}{3 * (Q^1 + Q^2 + Q^3 + T^1 + T^2 + T^3)} \times 100\%$$

Fig S7. ^{29}SI-NMR spectrum of AUP@SiOx and AUP@SiOx-184.

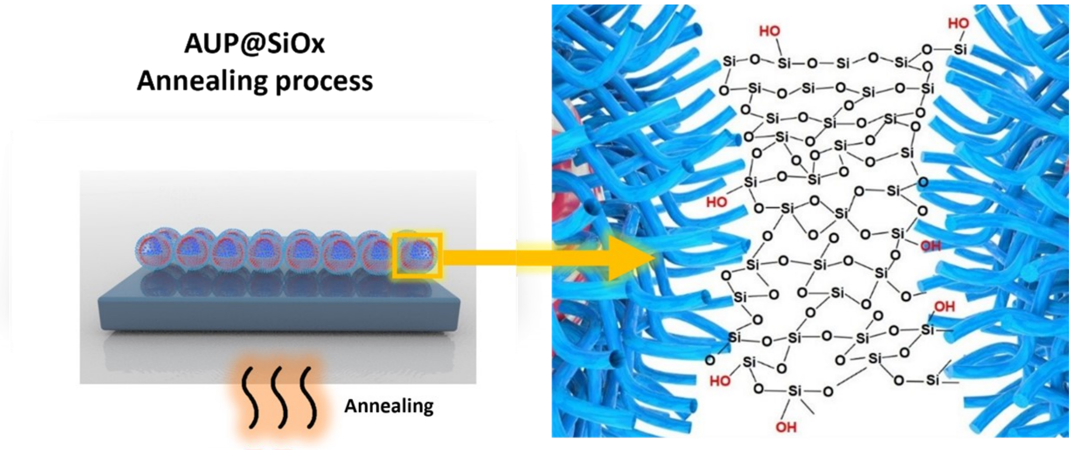


Fig S8. Schematic diagram of the film fabrication mechanism using a uniform AUP@SiOx sol-gel**.**

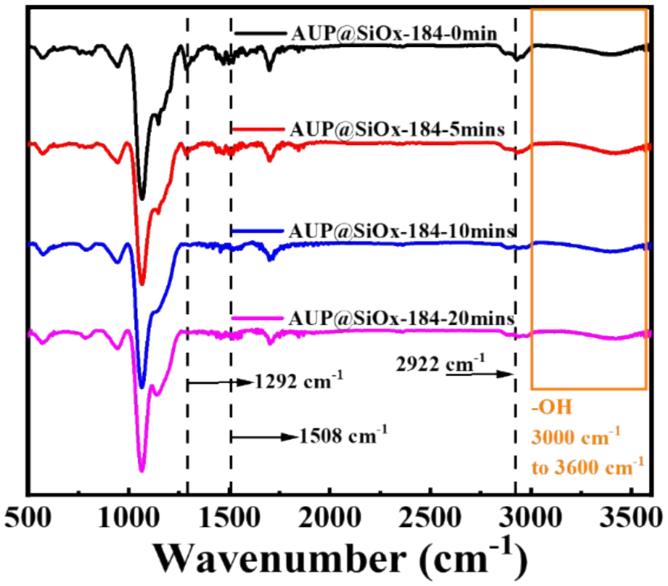


Fig. S9 FTIR spectra of AUP@SiOx films cured under different Annealing temperature.

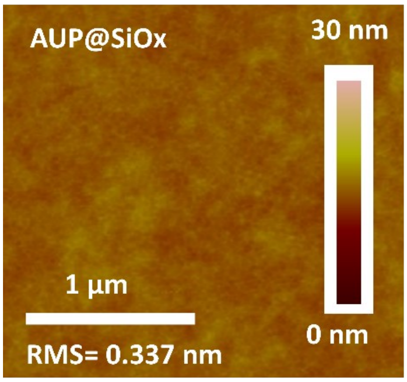


Figure S10. AFM image of AUP @SiOx.

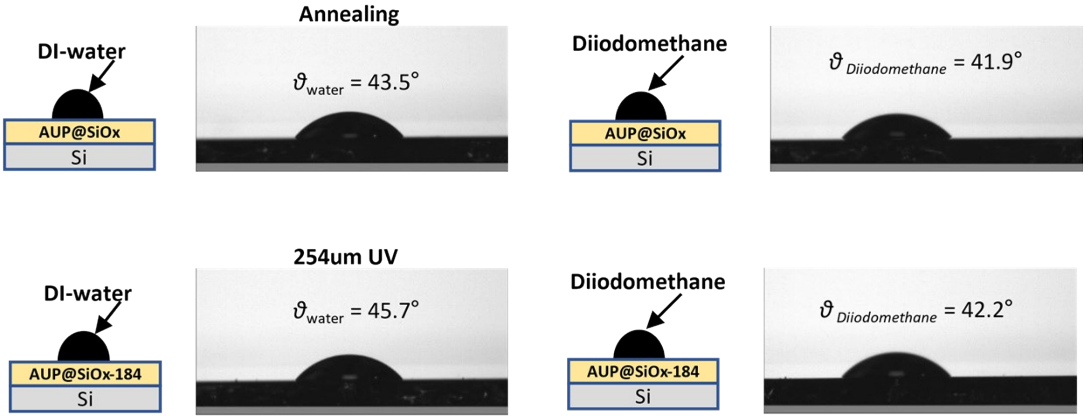


Fig. S11. Contact angle of the dielectric layer after curing of AUP@SiOx and AUP@SiOx-184.

Table S2. The surface tension of the two dielectric layers

Dielectric type	Contact Angle [º]		γ_s^p	γ_s^d	γ_s
	Water	Diiodomethane	[mJ m^{-2}]	[mJ m^{-2}]	[mJ m^{-2}]
AUP@SiOx	43.5	41.8	28.6	27.0	55.6
AUP@SiOx-184	45.7	42.2	27.0	27.2	54.2

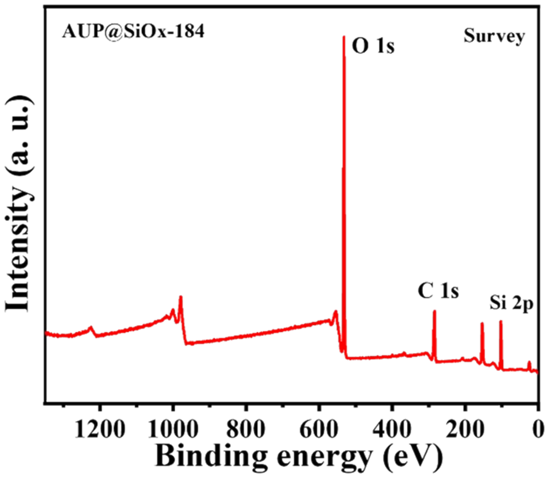


Fig S12. AUP@SiOx-184 full scan XPS spectra.

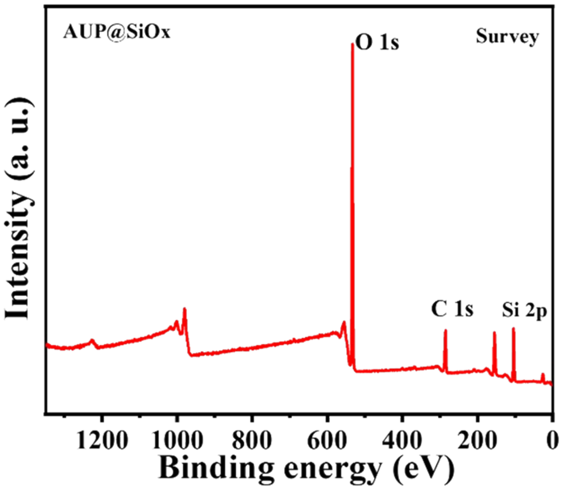


Fig S13. AUP@SiOx full scan XPS spectra.

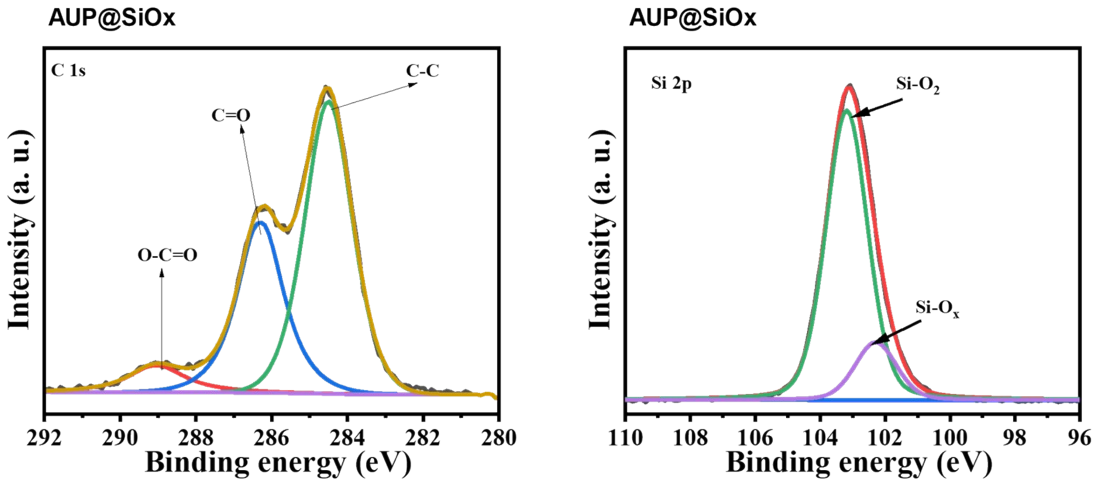


Fig S14. AUP@SiOx-184 C 1s and Si 2p XPS spectra.

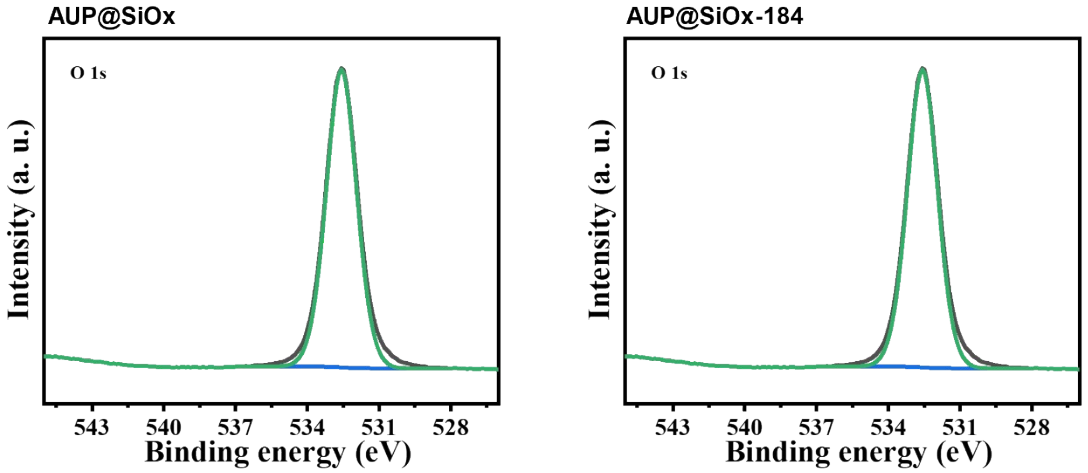


Fig S15. AUP@SiOx and AUP@SiOx-184 O 1s XPS spectra.

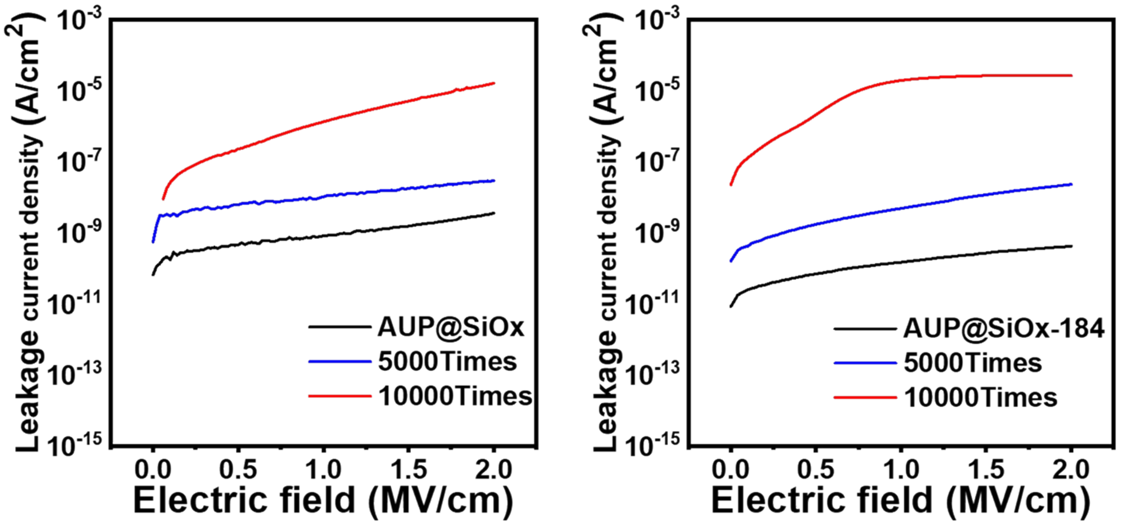


Fig S16. Leakage current stability under folding of AUP@SiOx and AUP@SiOx-184 MIM dielectric layers.

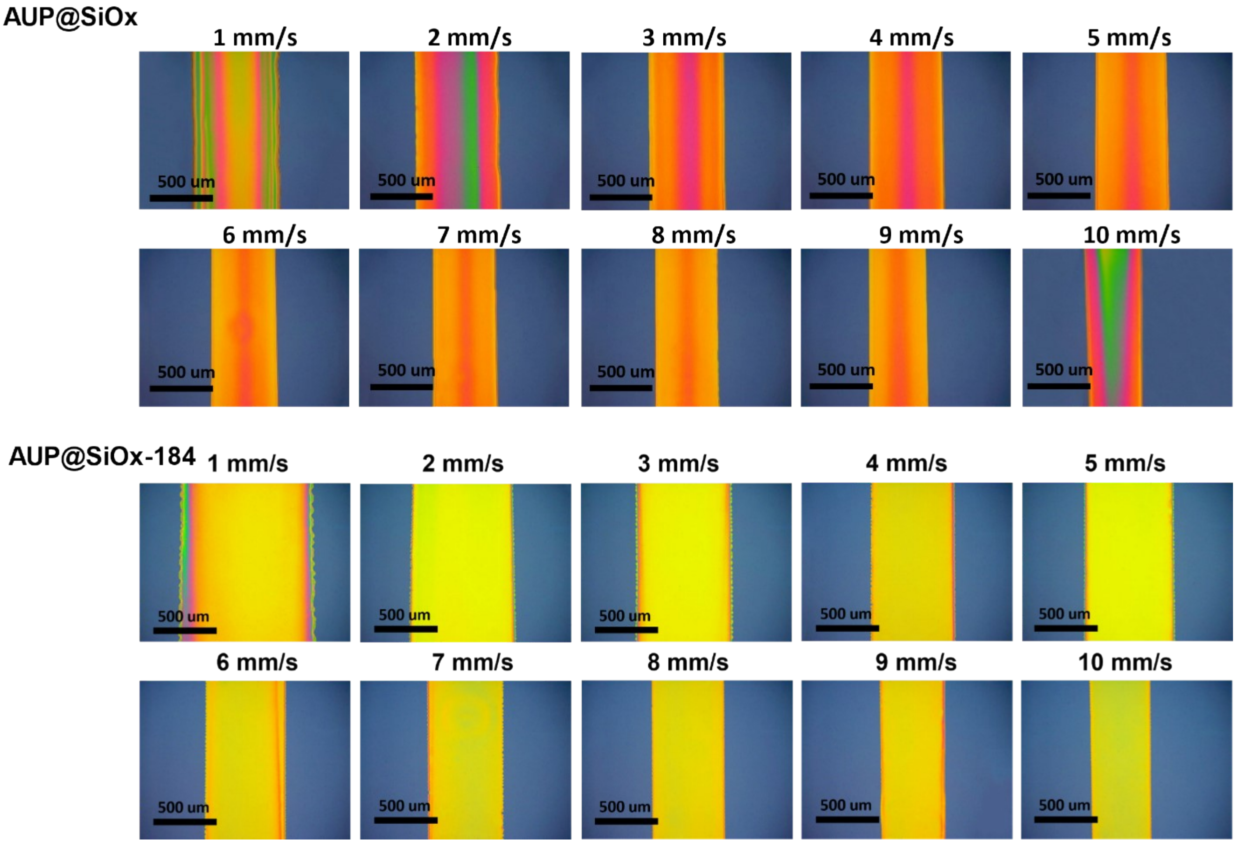


Fig. S17. EHD printed AUP@SiOx and AUP@SiOx-184 dielectric films at different printing speeds.

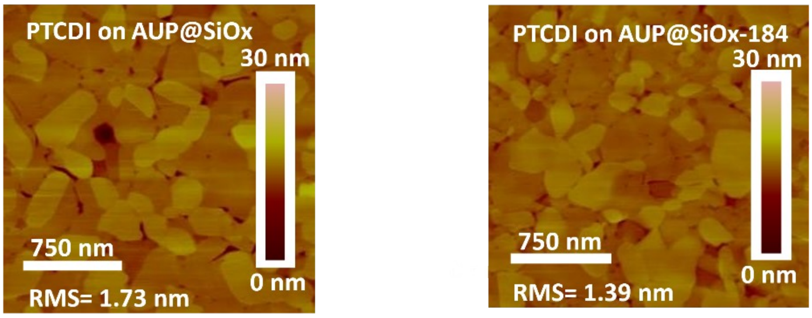


Fig. S18. AFM image for AUP@SiOx dielectric film (b) and AUP@SiOx dielectric film (e) with 50-nm-thick PTCDI-C_8.

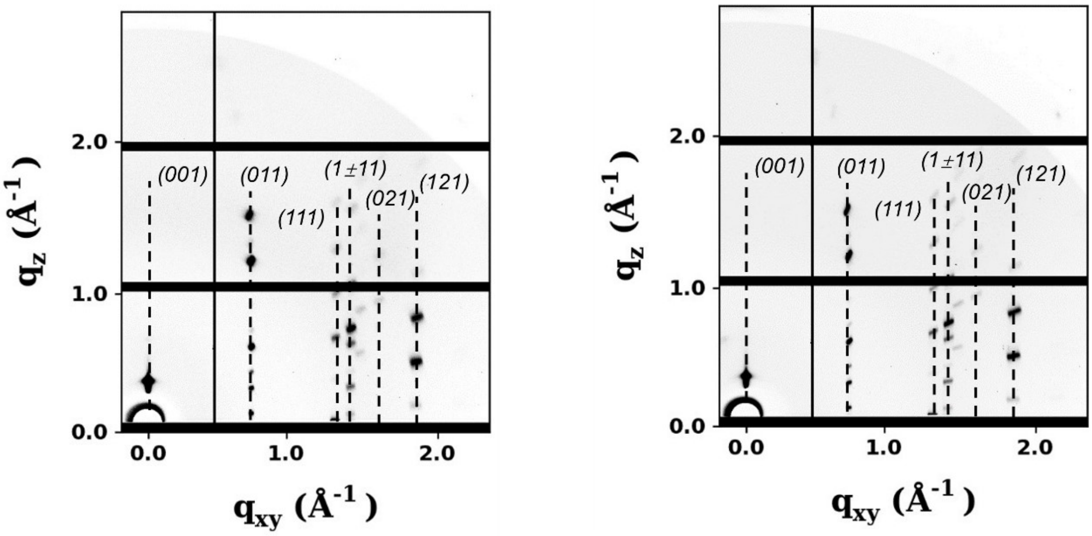


Fig. S19. The corresponding 2D-GIXD images of PTCDI-C_8

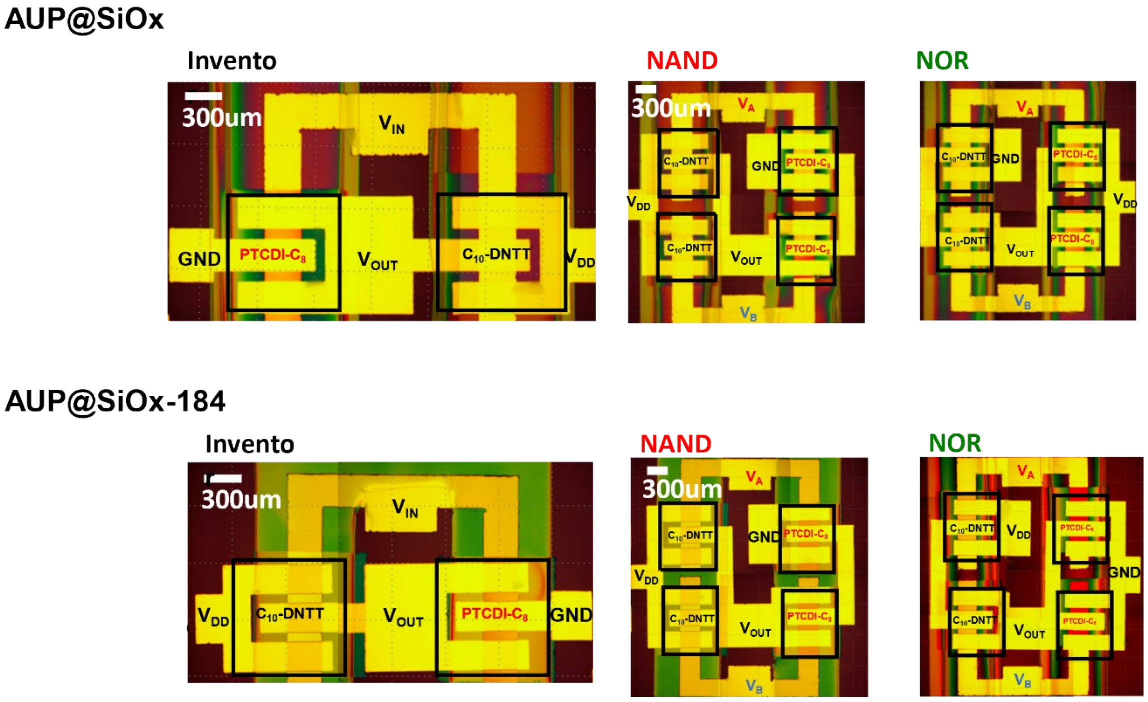


Fig. S20. OM image of NOT, NAND and NOR gates with C_{10}-DNTT and PTCDI-C_8 active layers with AUP@SiOx and AUP@SiOx-184 dielectric layers on the SiO_2 wafer has been prepared.

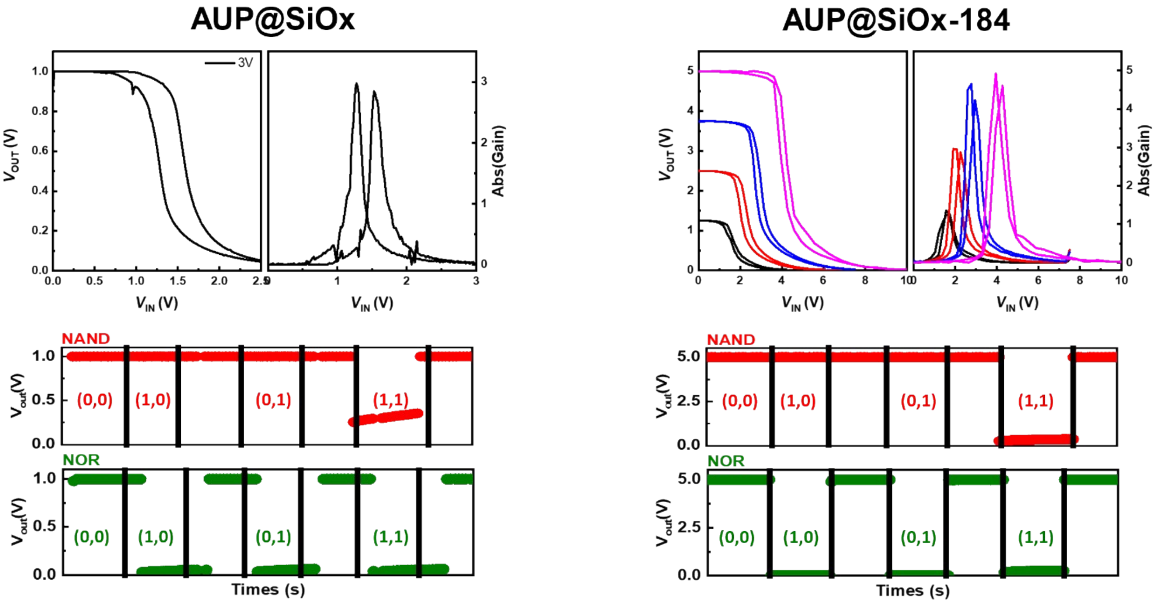


Fig. S21. NOT, NAND, and NOR logic gates printed by EHD, using AUP@SiOx for the dielectric layer (left) and AUP@SiOx-184 for the dielectric layer (right).

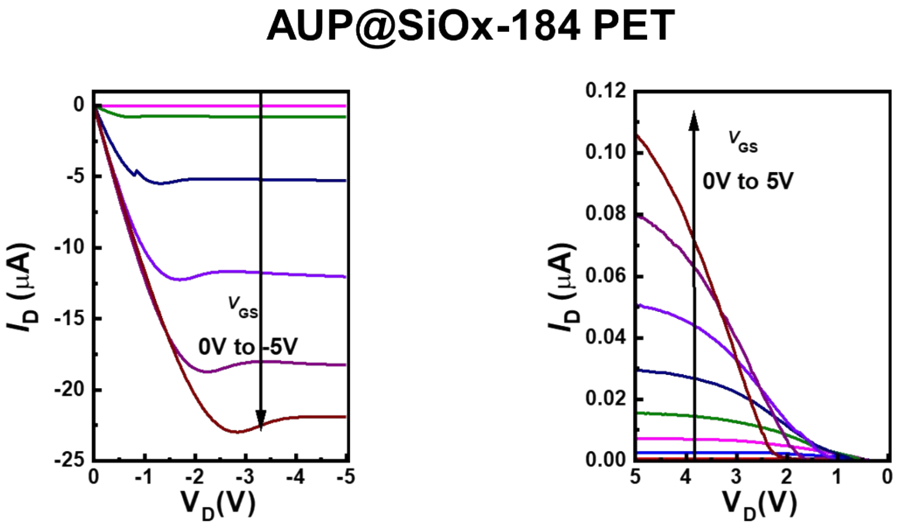


Fig. S22. Output characteristics of OTFTs prepared using EHD printing at PET films.