Supporting Information

Kinetic Swelling-Driven PTG Interlayers for Enhanced Crystallinity and Charge Transport in Polymer OECTs

Jeong Hye Song[a,†], *Yonghee Kim*[a,b,†], *Hocheon Yoo*[c], *Eun Kwang Lee*[a,*]

[a] Department of Chemical Engineering, Pukyong National University, Busan 48513, Republic of Korea

[b] Department of Chemical Engineering, Pohang University of Science and Technology (POSTECH), Pohang, Gyeongbuk 37673, Republic of Korea

[c] Department of Electronic Engineering, Hanyang University, 222 Wangsimni-ro, Seoul 04763, Republic of Korea

[†]These authors equally contributed to this work.

KEYWORDS.

Organic Electrochemical Transistors; PTG film; Kinetic Swelling; Crystallinity; Neuromorphic Devices

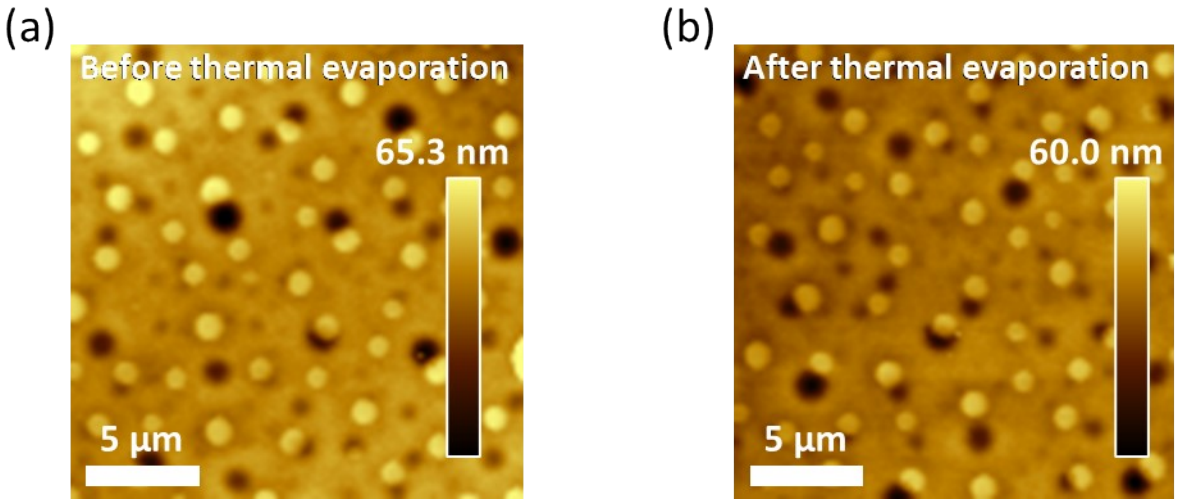

Figure S1. AFM images of the PTG film (a) before and (b) after thermal evaporation of Au electrodes under identical vacuum and temperature conditions. No significant morphology changes were observed.

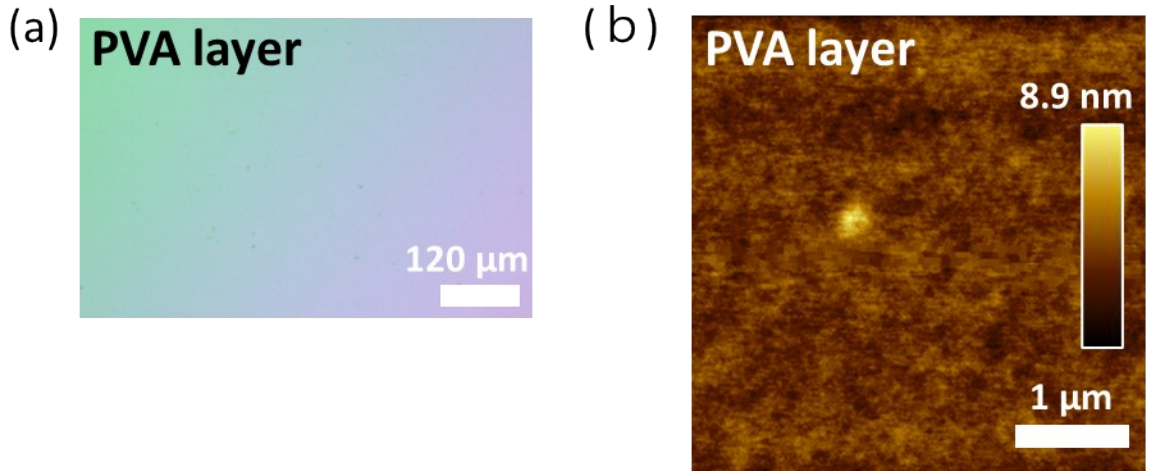

Figure S2. Analysis of the surface of PVA layer. (a) OM image and (b) the AFM height image of PVA film layer.

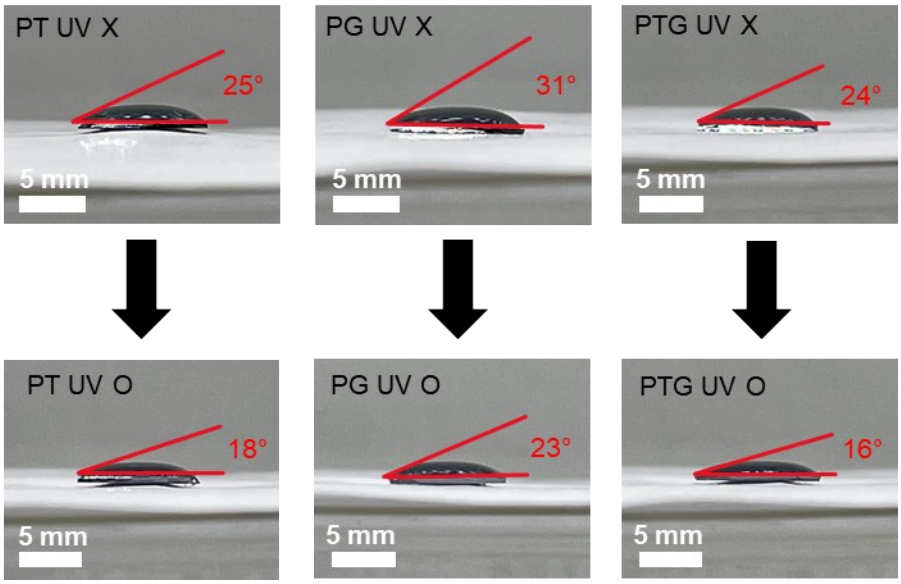

Figure S3. The 20 μL water contact angle results on PVA, PT and PTG layer.

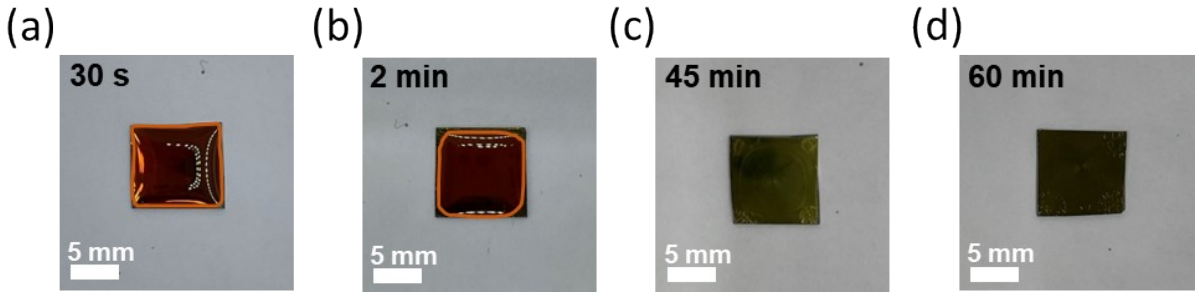

Figure S4. Pictures of P3HT solution on PTG film showing swelling property for wetting time of (a) 30 s, (b) 2 min, (c) 45 min and (d) 60 min.

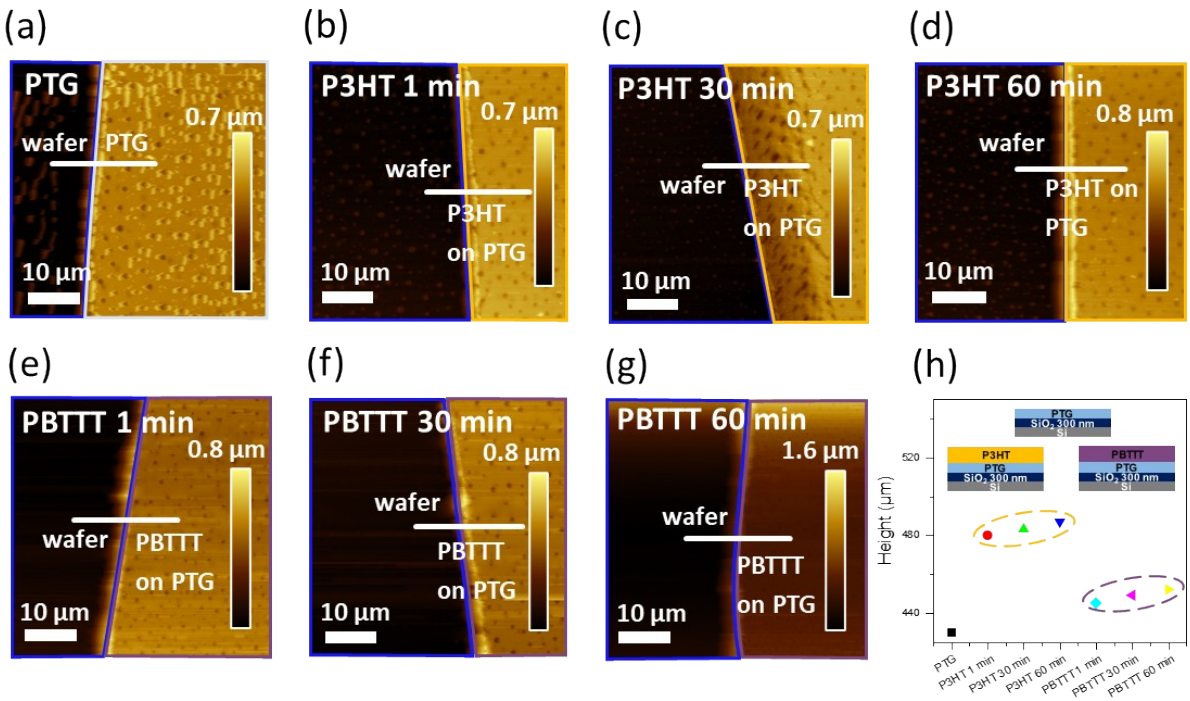

Figure S5. AFM height images of (a) pristine PTG film, (b–d) P3HT-coated, and (e–g) PBTTT-coated PTG films after 1, 30 and 60 min of wetting. Each image shows the height difference between the coated film and the bare wafer, used to estimate the relative thickness increase due to polymer absorption and film formation. (h) Summary of thickness extracted from (a–g).

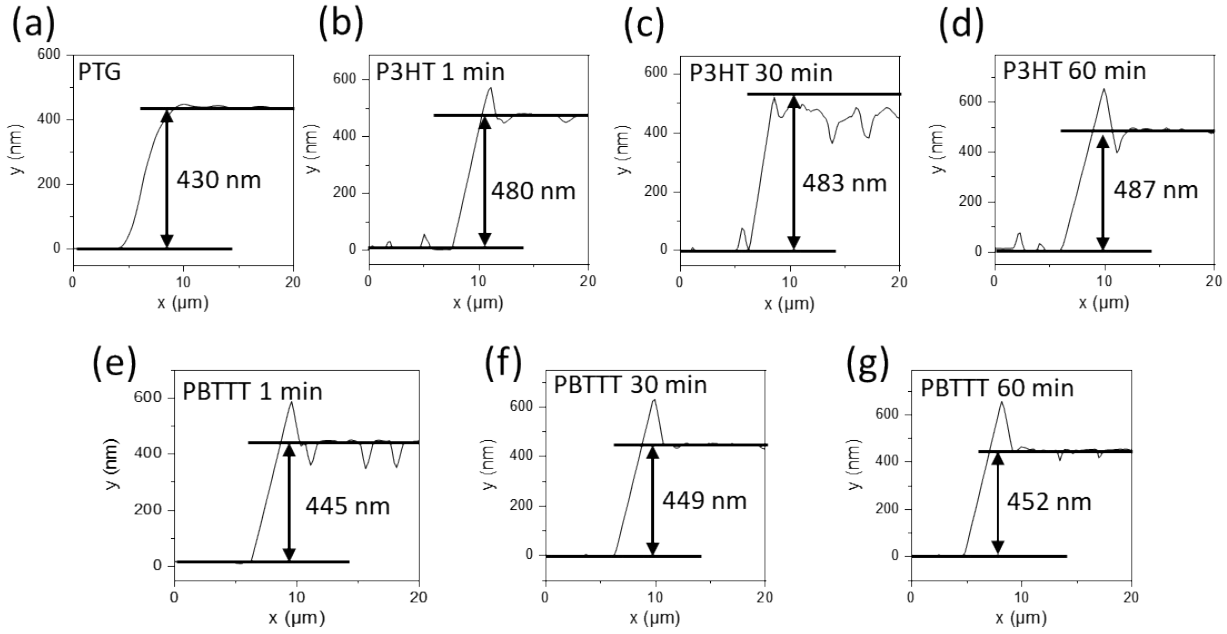

Figure S6. AFM height profiles of (a) pristine PTG film, (b-d) P3HT-coated and (e-f) PBTTT-coated PTG films after 1, 30 and 60 min of wetting, respectively.

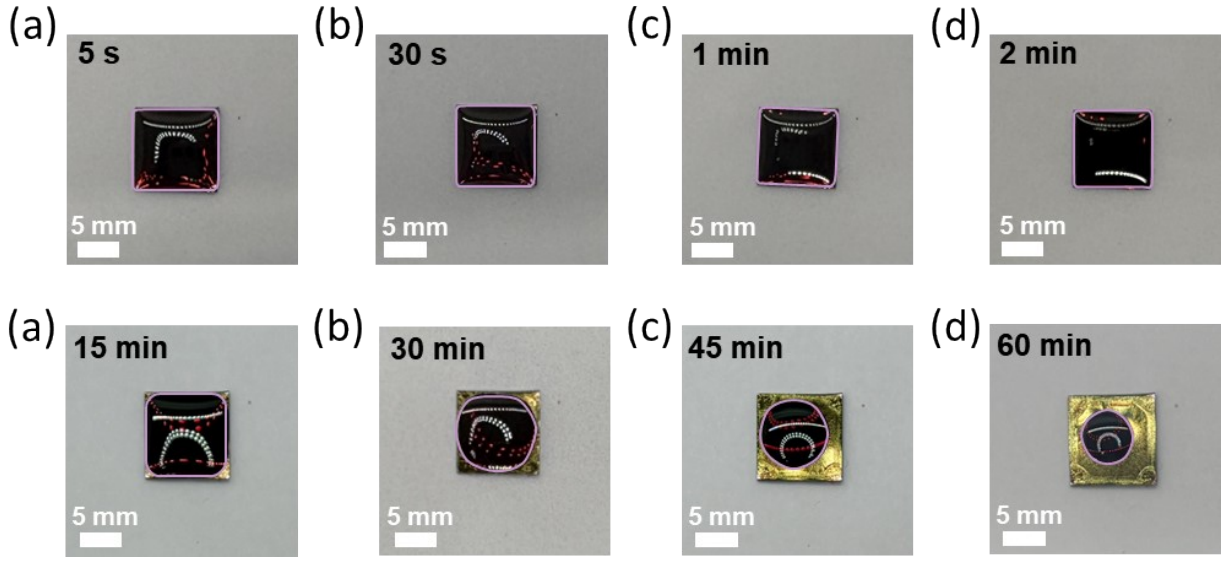

Figure S7. Pictures of PBTTT solution on PTG film showing swelling property for wetting time of (a) 5 s, (b) 30 s, (c) 1 min (d) 2 min, (e) 15 min, (f) 30 min, (g) 45 min and (h) 60 min.

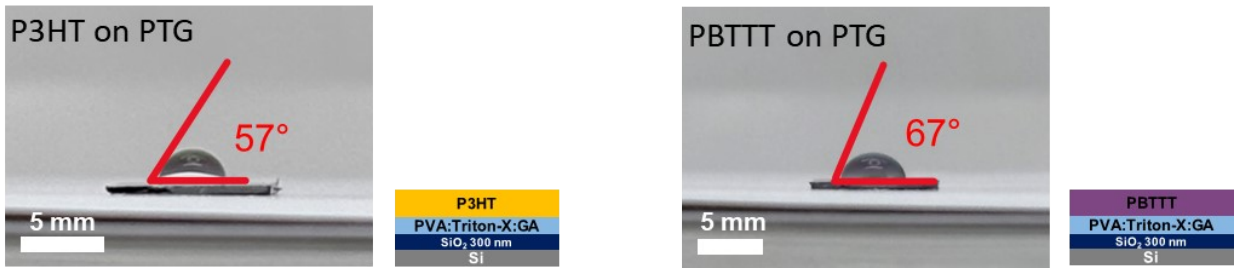

Figure S8. The 20 μL water contact angle results on P3HT and PBTTT on PTG layer.

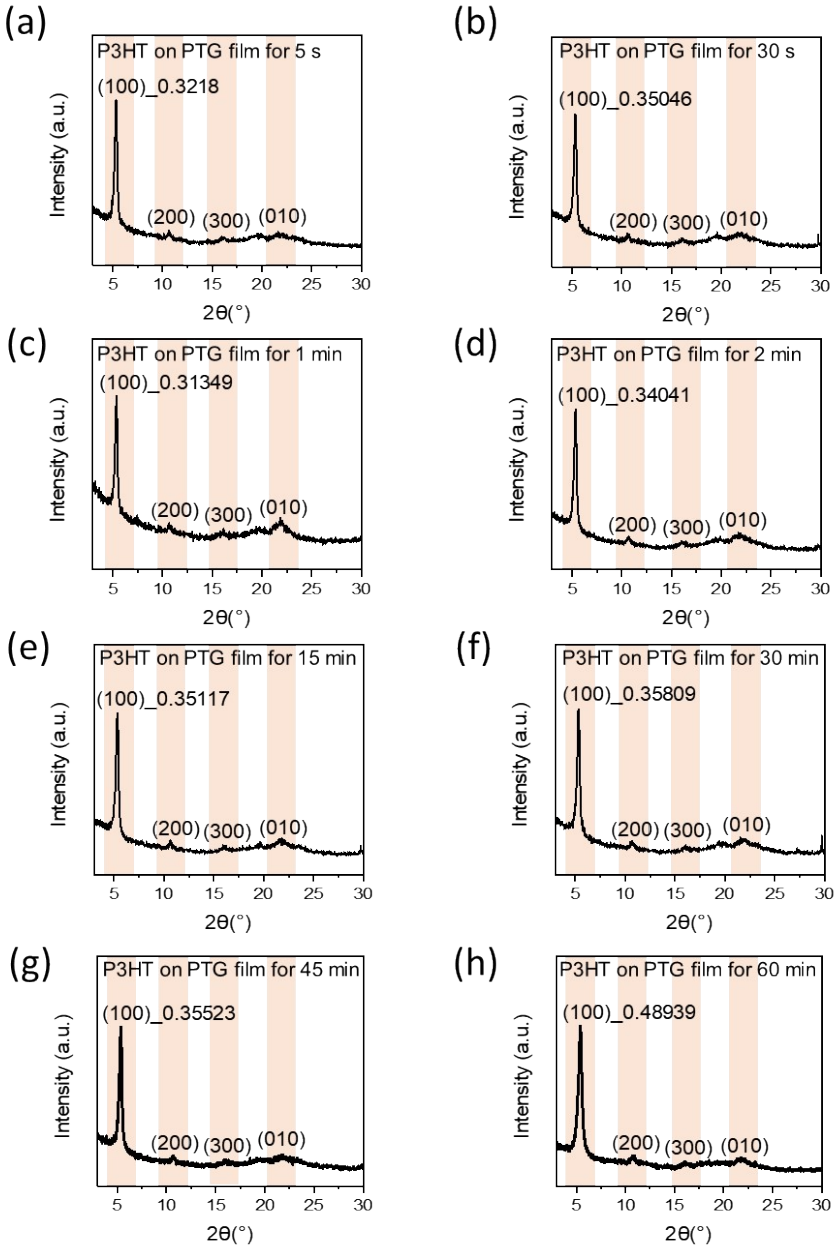

Figure S9. XRD spectra of P3HT crystal on PTG layer with each wetting waiting time: (a) 5 s, (b) 30 s, (c) 1 min, (d) 2 min, (e) 15 min, (f) 30 min, (g) 45 min and (h) 60 min.

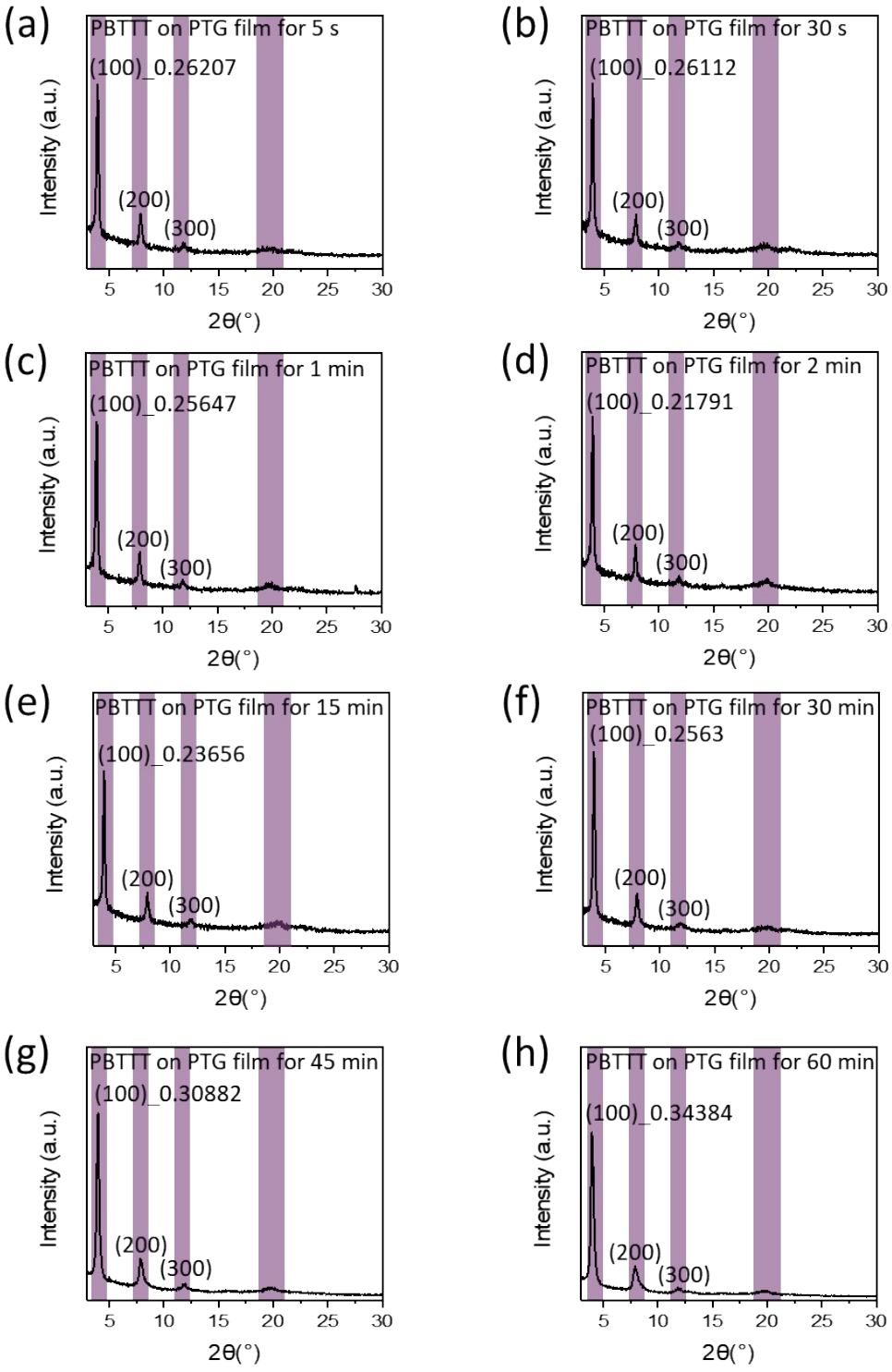

Figure S10. XRD spectra of PBTTT on PTG layer with each wetting waiting time: (a) 5 s, (b) 30 s, (c) 1 min, (d) 2 min, (e) 15 min, (f) 30 min, (g) 45 min and (h) 60 min.

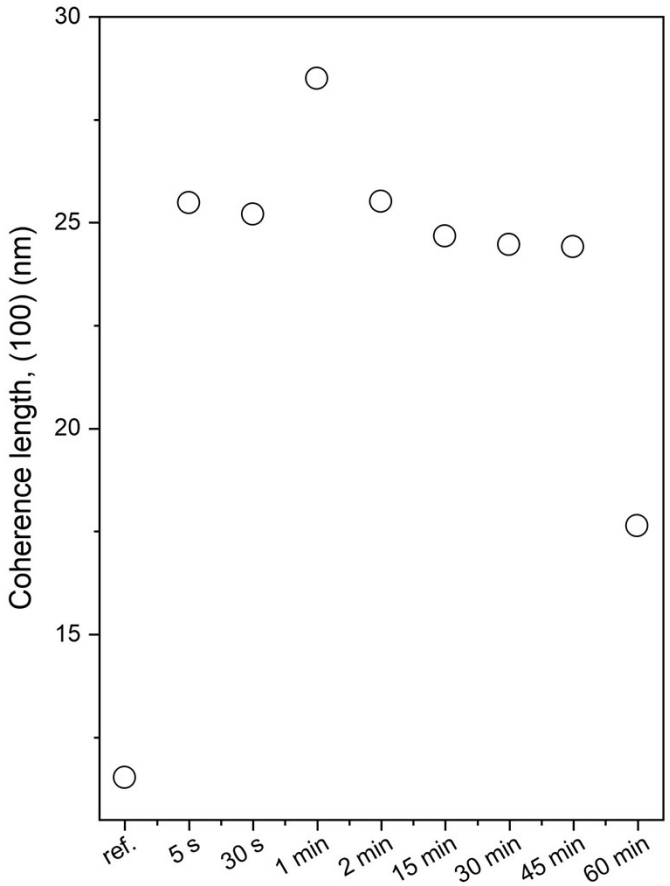

Figure S11. Calculated coherence length from FWHM of (100) peaks in XRD spectra of the P3HT polymer crystalline structures according to wetting time.

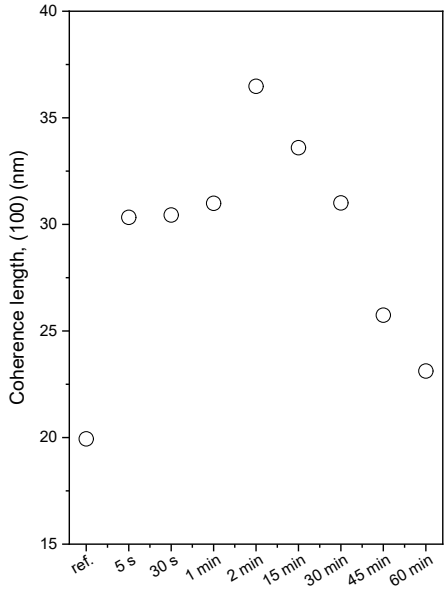

Figure S12. Calculated coherence length from FWHM of (100) peaks in XRD spectra of the PBTTT polymer crystalline structures according to wetting time.

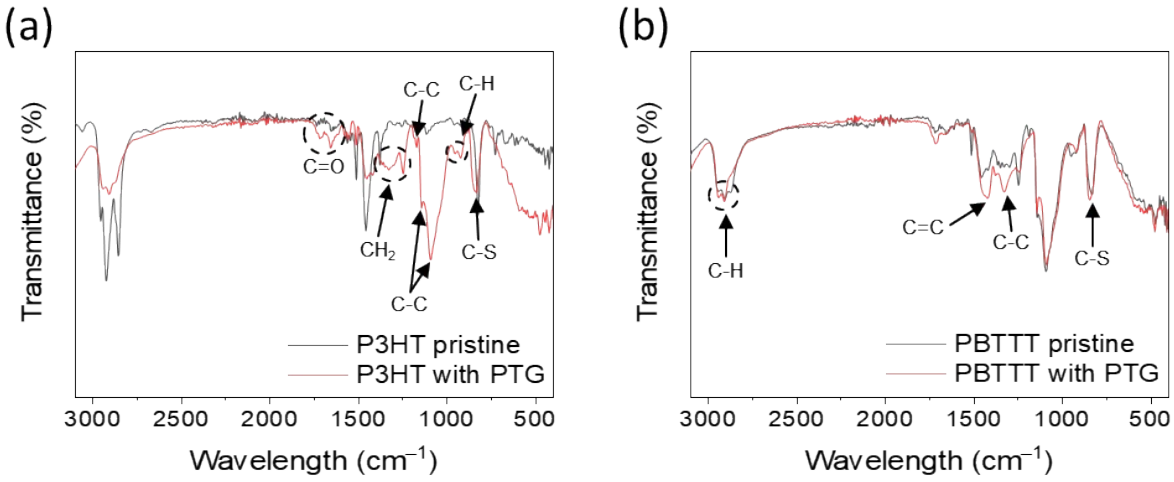

Figure S13. FT-IR spectra of (a) P3HT and (b) PBTTT films without and with PTG supporting layer.

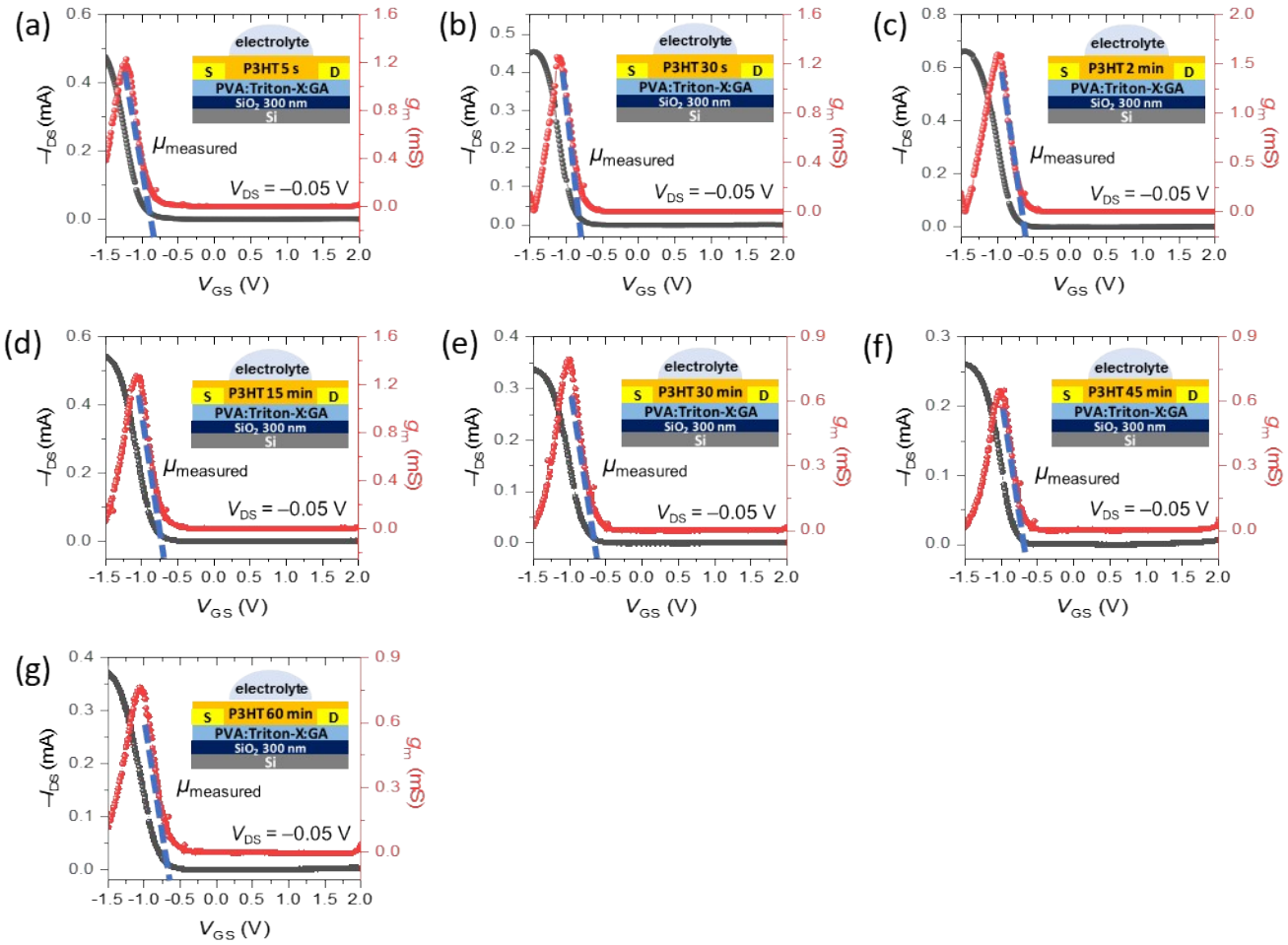

Figure S14. Transfer curves of P3HT OECTs on PTG film according to wetting time: (a) 5 s, (b) 30 s, (c) 2 min, (d) 15min, (e) 30min, (f) 45min and (g) 60min. The transfer curves were acquired by scanning V_{GS} from −1.5 to 2 V in 0.01 V increments with a constant V_{DS} of −0.05 V.

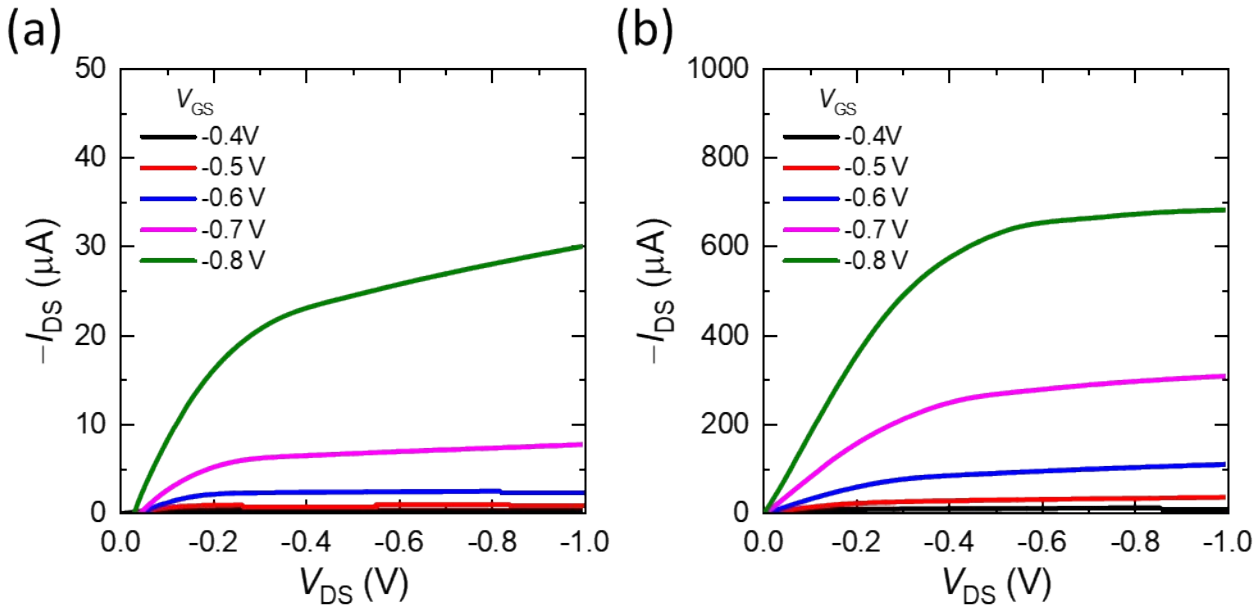

Figure S15. output curves of OECTs based on pristine P3HT and P3HT crystals on PTG film for wetting time of 1 min. The output curves were obtained by adjusting V_{DS} from −1 to 0 V in 0.01 V increments while applying V_{GS} values of −0.4, −0.5, −0.6, −0.7, and −0.8 V.

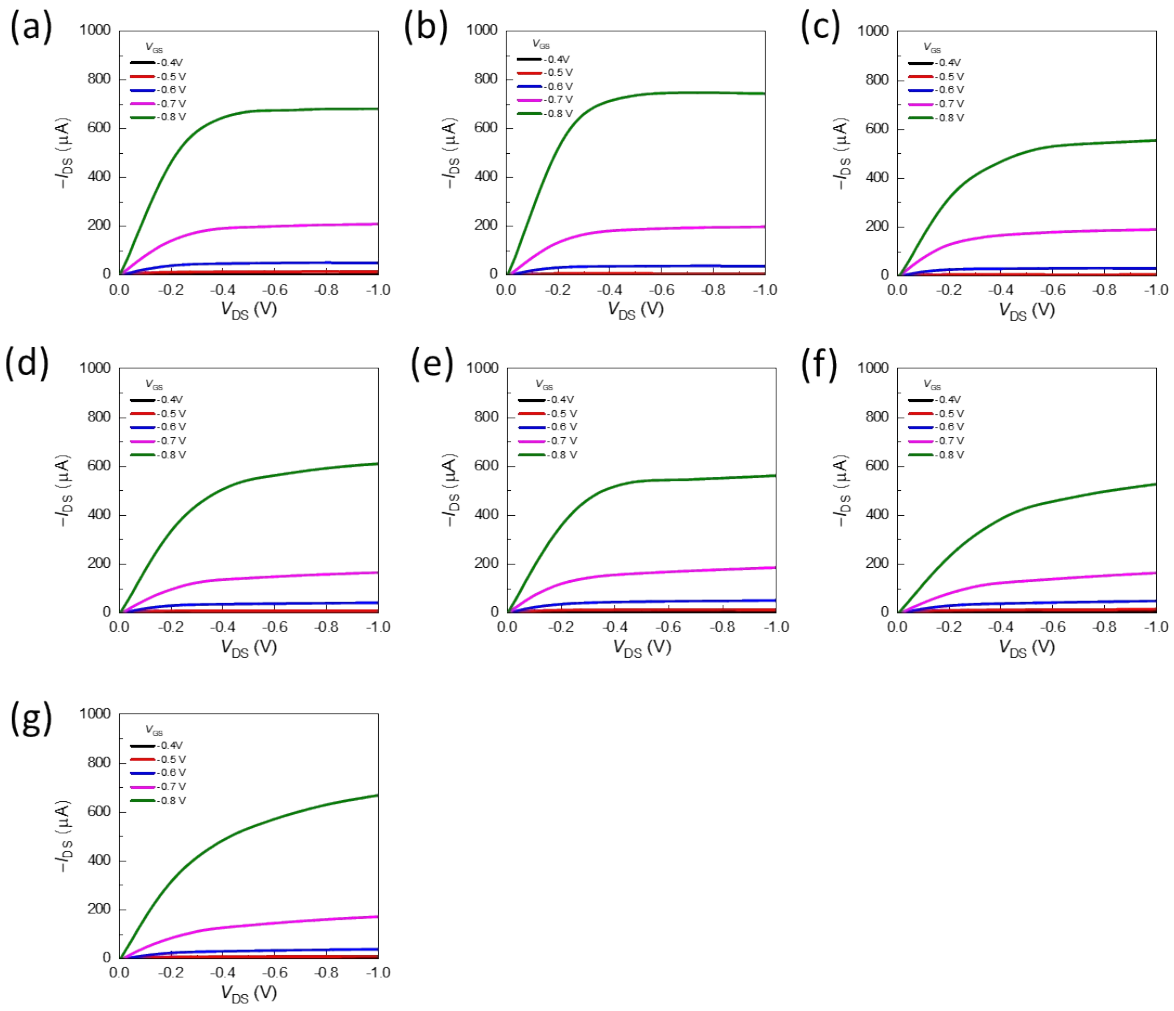

Figure S16. Output curves of OECTs based on P3HT crystal on PTG film for wetting time: (a) 5 s and (b) 30 s, (c) 2 min, (d) 15 min, (e) 30 min, (f) 45 min and (g) 60min. The output curves were obtained by adjusting V_{DS} from −1 to 0 V in 0.01 V increments while applying V_{GS} values of −0.4, −0.5, −0.6, −0.7, and −0.8 V.

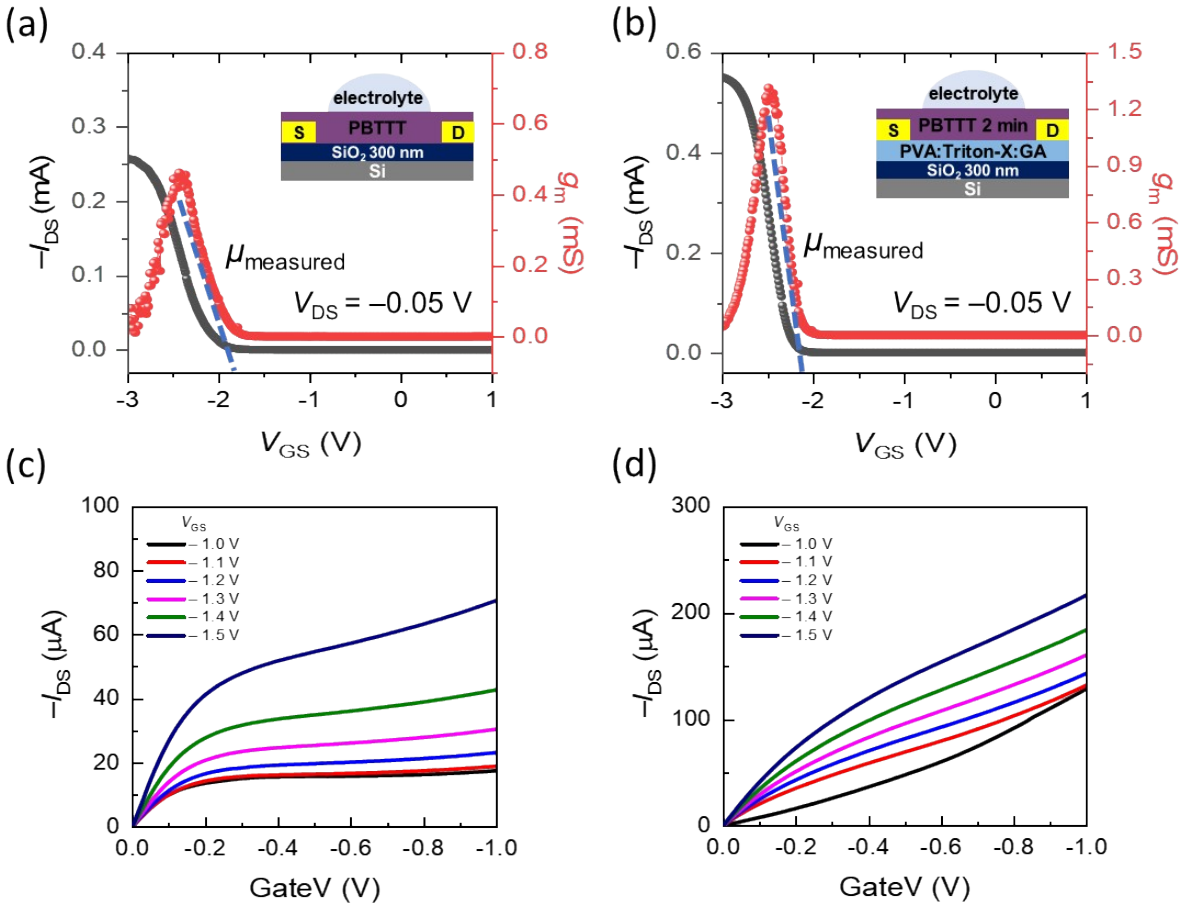

Figure S17. OECT transfer curves of (a) PBTTT formed on SiO_2 and (b) on PTG film for 2 min wetting time. Output curves of OECTs based (c) on pristine PBTTT and (d) PBTTT on PTG film for wetting time of 2 min.

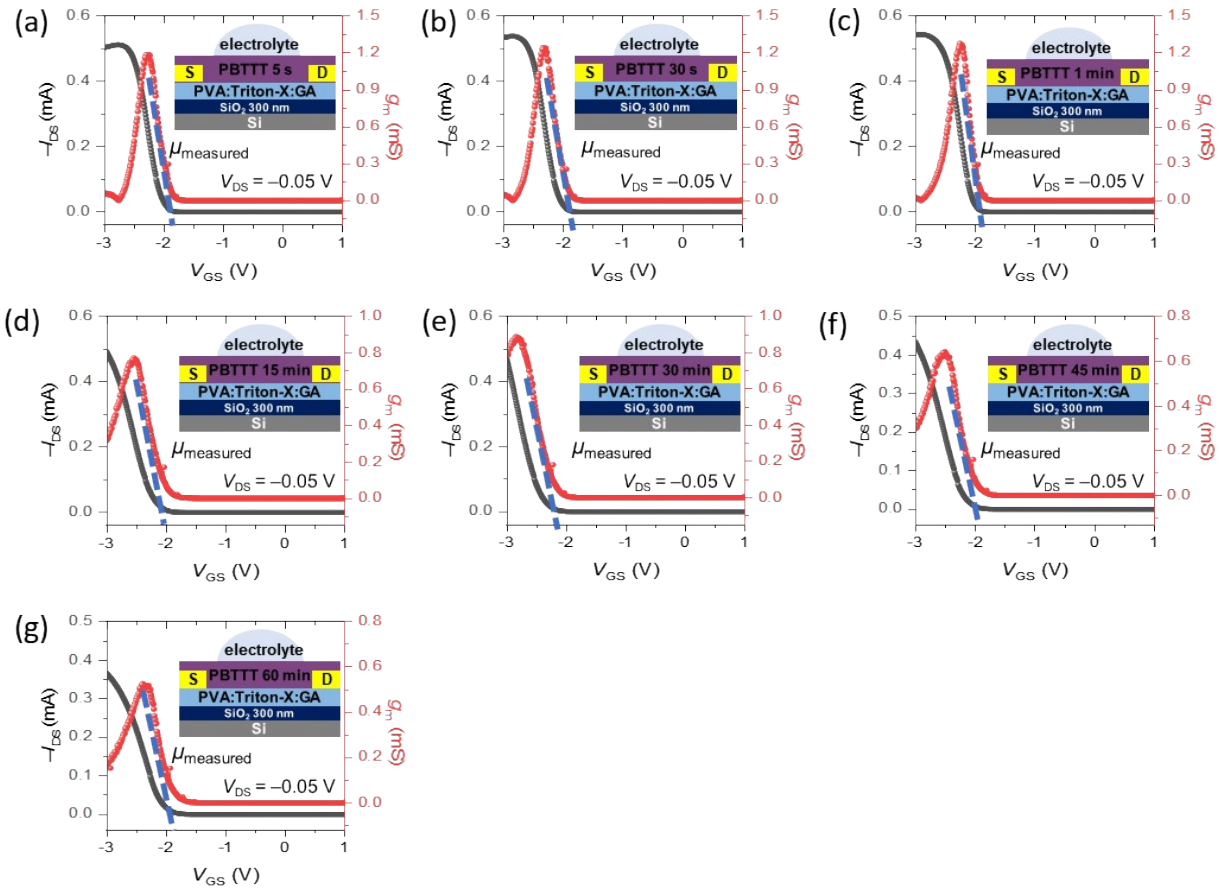

Figure S18. Transfer curves of PBTTT on PTG film OECTs in each wetting time: (a) 5 s, (b) 30 s, (c) 1 min, (d) 15min, (e) 30min, (f) 45min and (g) 60min.

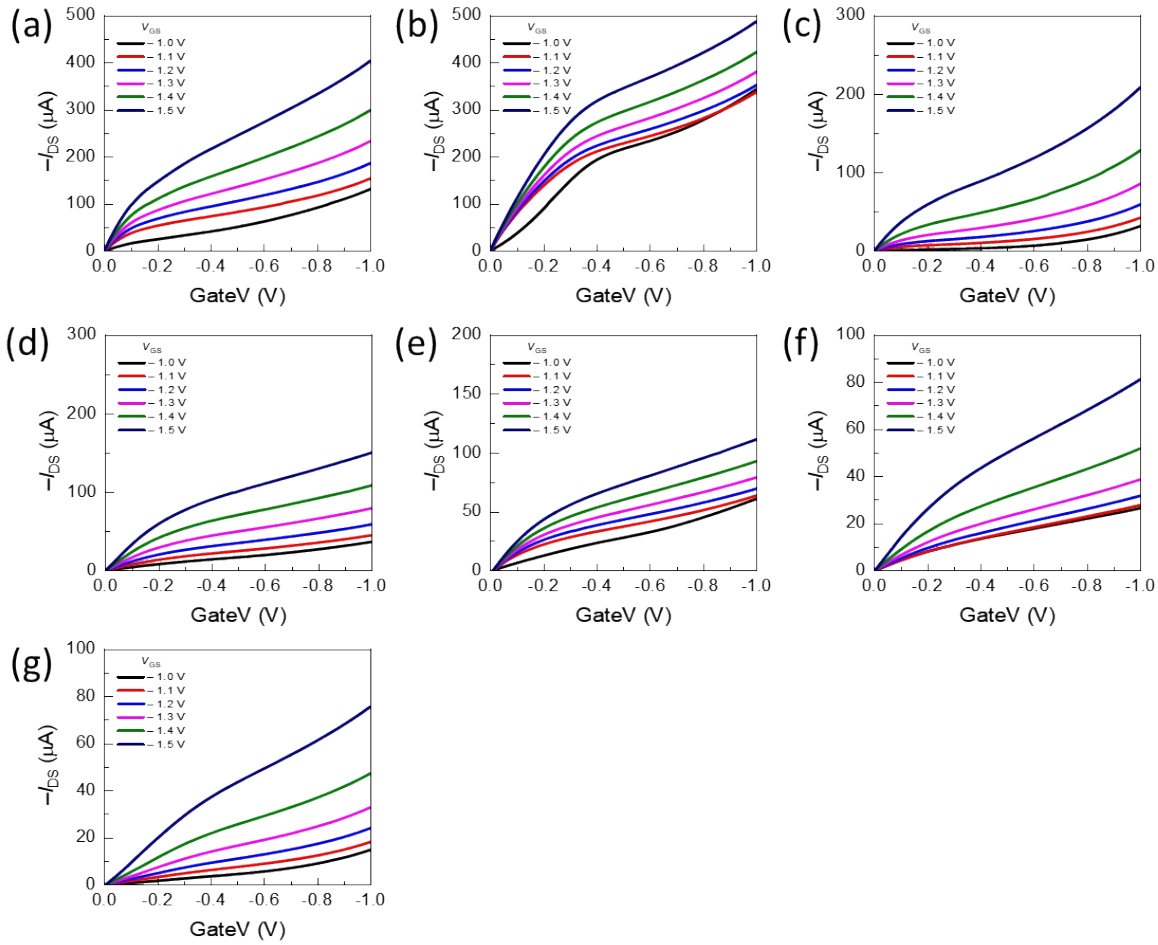

Figure S19. Output curves of OECTs with PBTTT on PTG film for each wetting time: (a) 5 s and (b) 30 s, (c) 1 min, (d) 15 min, (e) 30 min, (f) 45 min and (g) 60min.

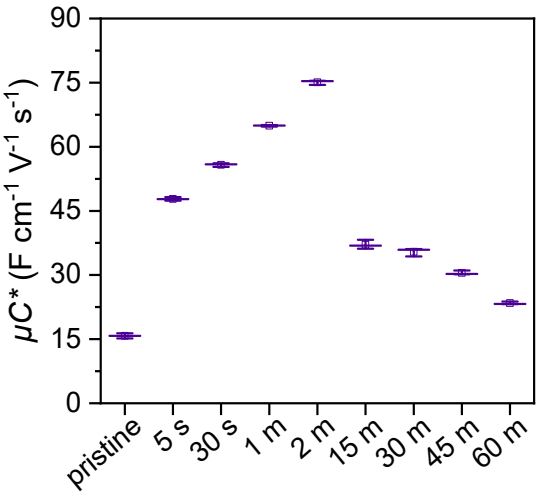

Figure S20. μC^* of PBTTT on SiO_2, and on PTG film as a function of wetting time (from 5 s to 60 min).

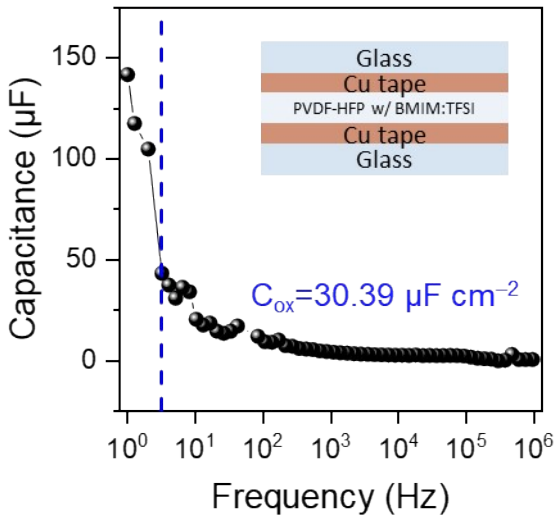

Figure S21. EIS measurement of the PVDF-HFP ion gel containing BMIM:TFSI.

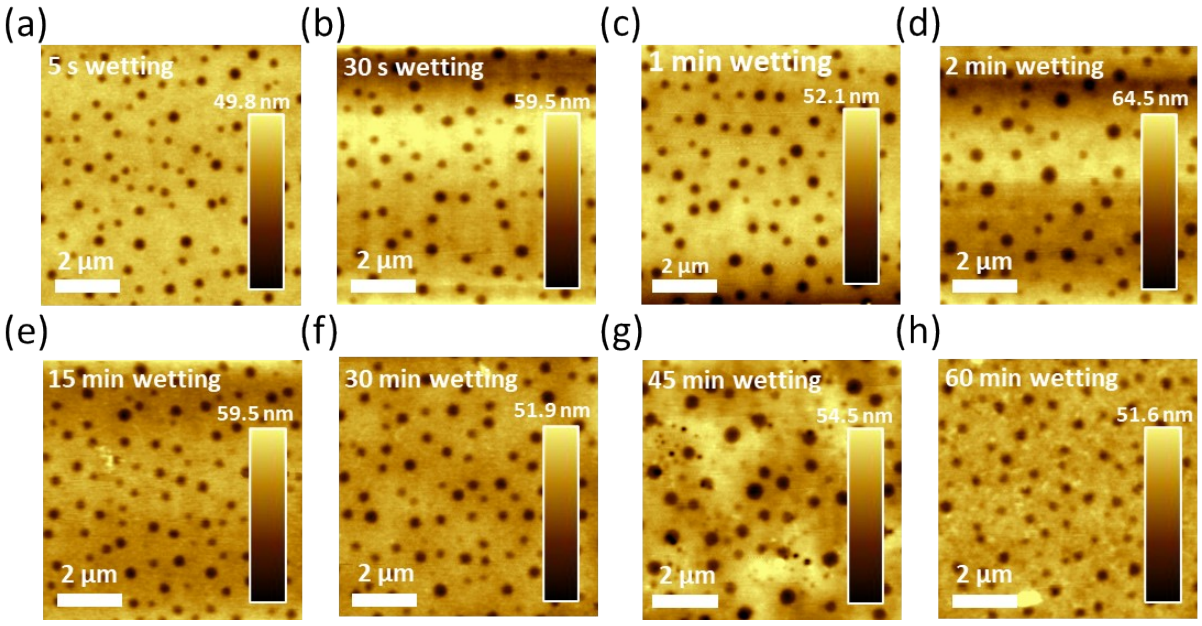

Figure S22. 10 × 10 µm AFM results for P3HT on PTG film for each wetting time condition: (a) 5 s and (b) 30 s, (c) 1 min, (d) 2 min, (e) 15 min, (f) 30 min, (g) 45 min and (h) 60 min.

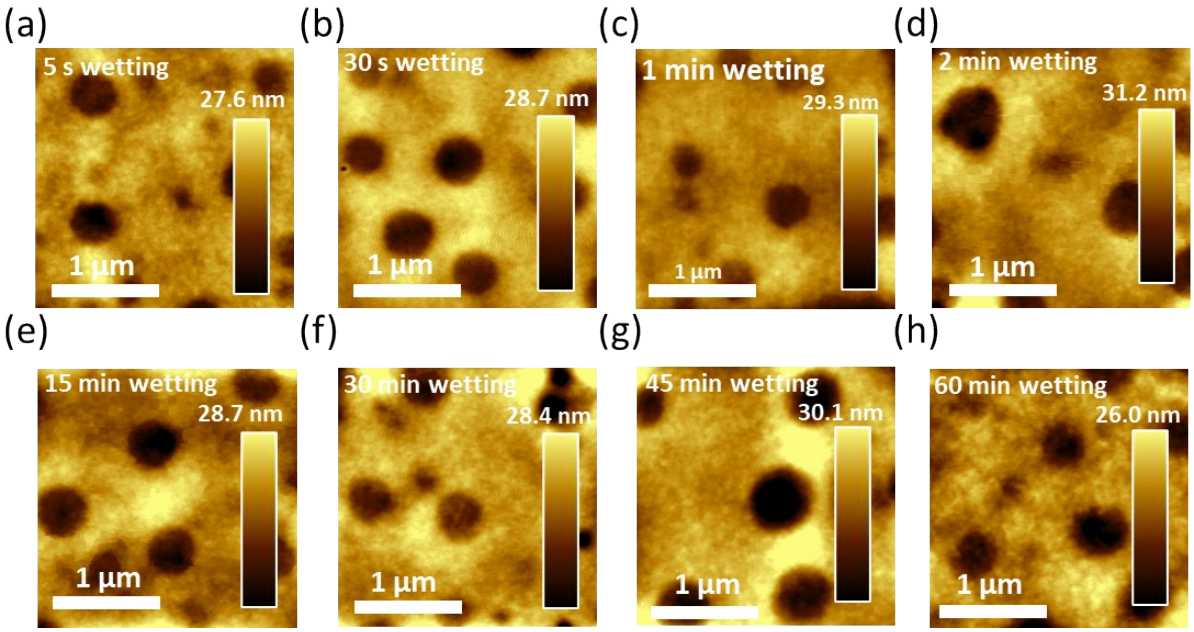

Figure S23. 3 × 3 µm AFM results for P3HT on PTG film for each wetting waiting time condition: (a) 5 s and (b) 30 s, (c) 1 min, (d) 2 min, (e) 15 min, (f) 30 min, (g) 45 min and (h) 60 min.

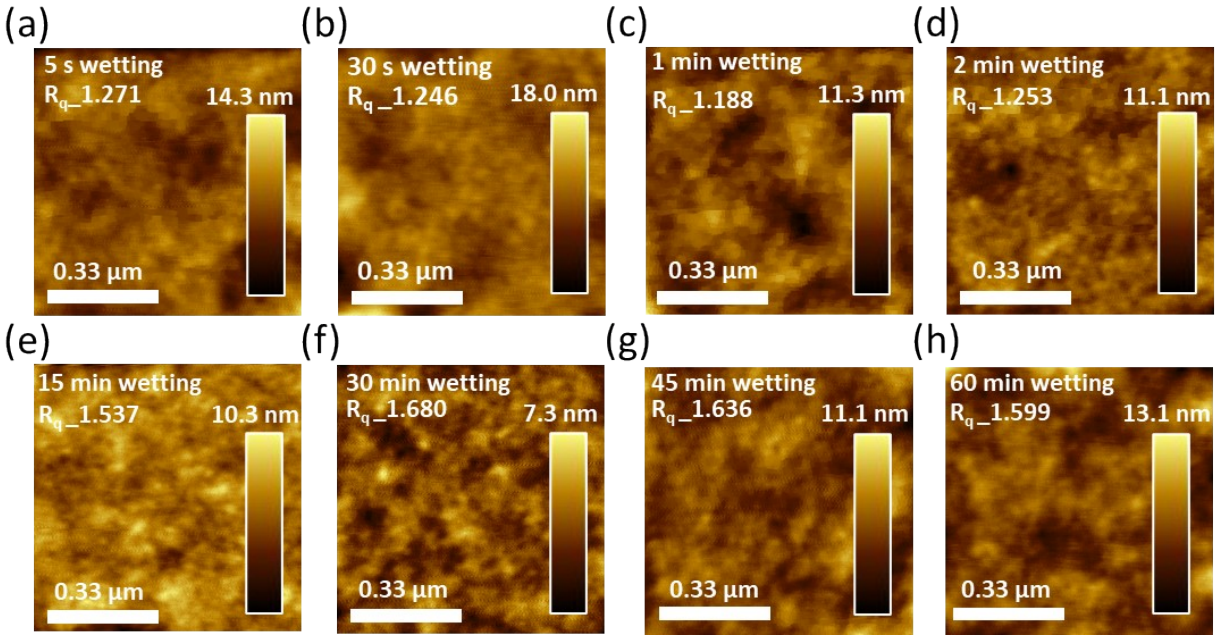

Figure S24. 1 × 1 µm AFM results and R_q for P3HT on PTG film for each wetting time condition: (a) 5 s and (b) 30 s, (c) 1 min, (d) 2 min, (e) 15 min, (f) 30 min, (g) 45 min and (h) 60 min.

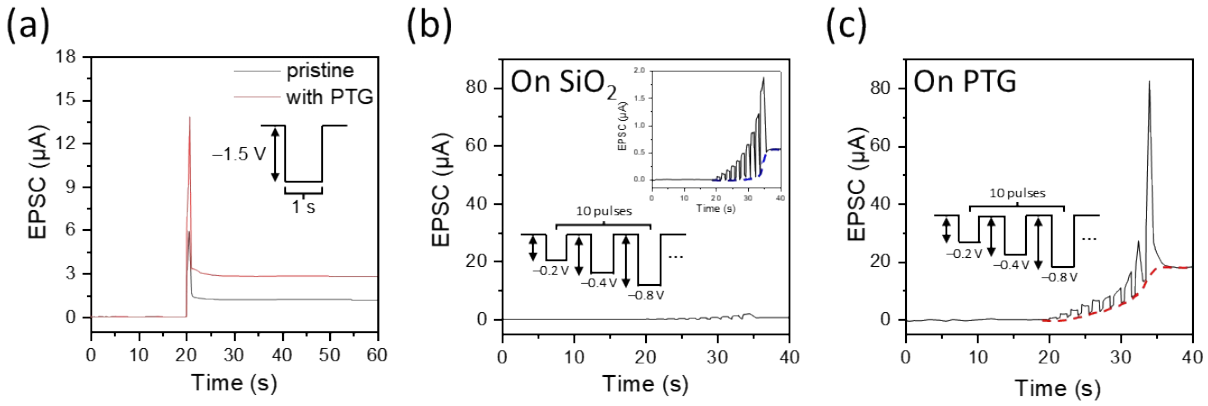

Figure S25. (a) EPSC of P3HT-based devices after single presynaptic spikes (V_{GS} = –1.5 V and V_{DS} = –1 V) on SiO_2 and PTG layer. Synaptic characteristics under variations in presynaptic pulse strength (from –0.2 V to –2.0 V in –0.2 V steps) for (b) SiO_2 and (c) PTG layer.

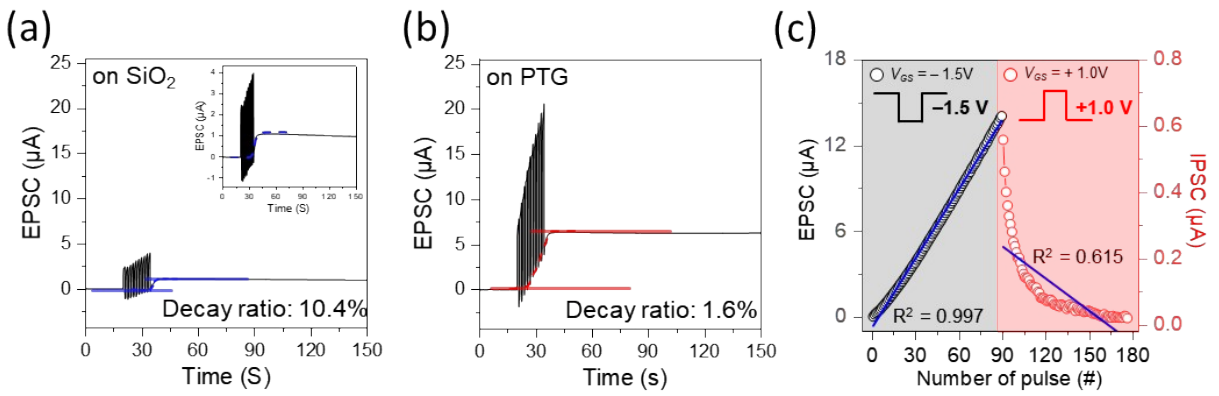

Figure S26. LTM properties P3HT OECT formed on (a) SiO_2 and (b) PTG layer up to 115 s after 10 pulses for each condition. (c) LTP/LTD characteristics of P3HT OECT formed on SiO_2 under the application of 90 potentiation pulses at – 1.5 V and 87 depression pulses at + 1.0 V.

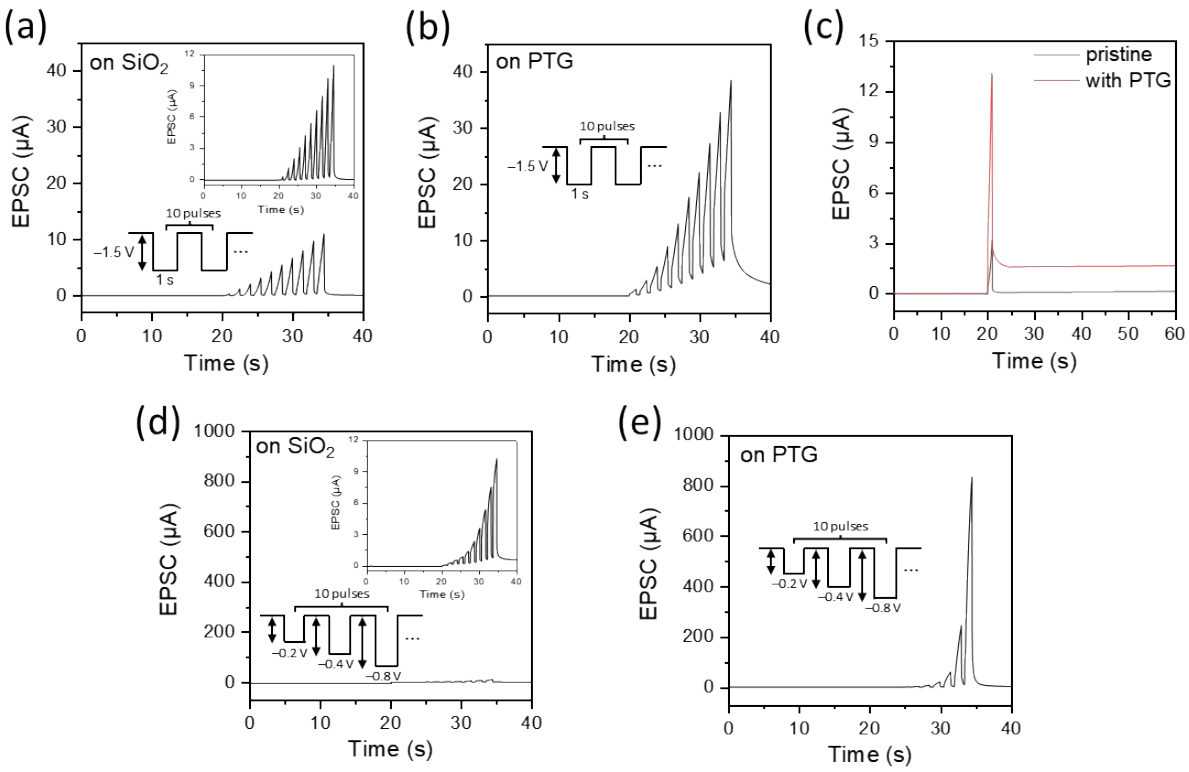

Figure S27. EPSC of PBTTT based devices observed after 10 presynaptic spikes (V_{GS} = –1.5 V, V_{DS} = –1 V and spike time interval (Δ t = 0.5 s) on (a) SiO_2 and (b) PTG layer. (c) EPSC of PBTTT based devices after single presynaptic spikes (V_{GS} = –1.5 V and V_{DS} = –1 V) on SiO_2 and PTG layer. Synaptic characteristics under variations in presynaptic pulse strength (from –0.2 V to –2.0 V in –0.2 V steps) for (d) SiO_2 and (e) PTG layer.

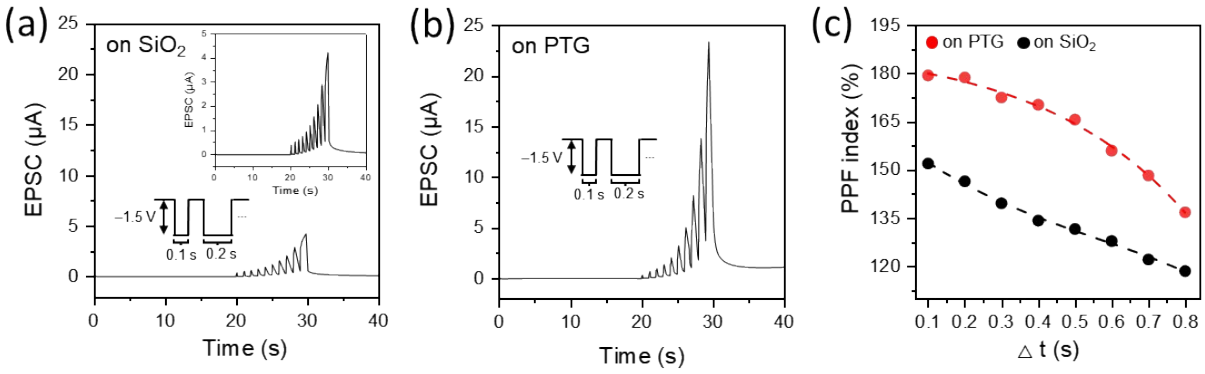

Figure S28. EPSC of each device as presynaptic spike widths increase from 0.1 s to 0.9 s in 0.1 s steps (V_{GS} = –1.5 V, V_{DS} = –1 V) on (a) SiO_2 and (b) PTG layer. (c) PPF index of OECT with PBTTT formed on SiO_2 (black) and PTG (red) layer, according to Δ t changed from 0.1 s to 0.8 s.

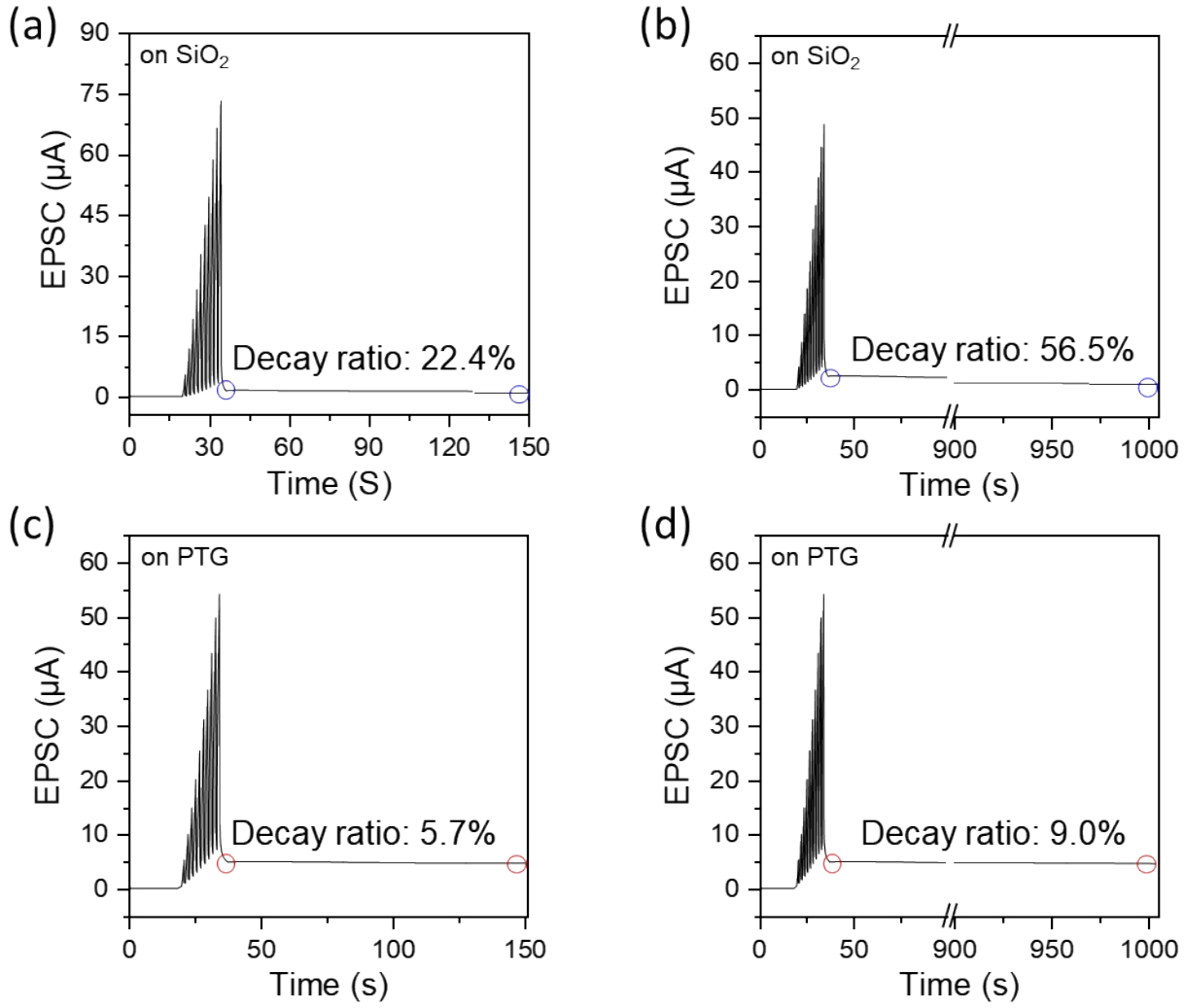

Figure S29. LTM properties PBTTT OECT formed on SiO_2 and PTG layer up to 115 s and 1005 s after 10 pulses for each condition: (a) and (b) SiO_2, (c) and (d) PTG layer.

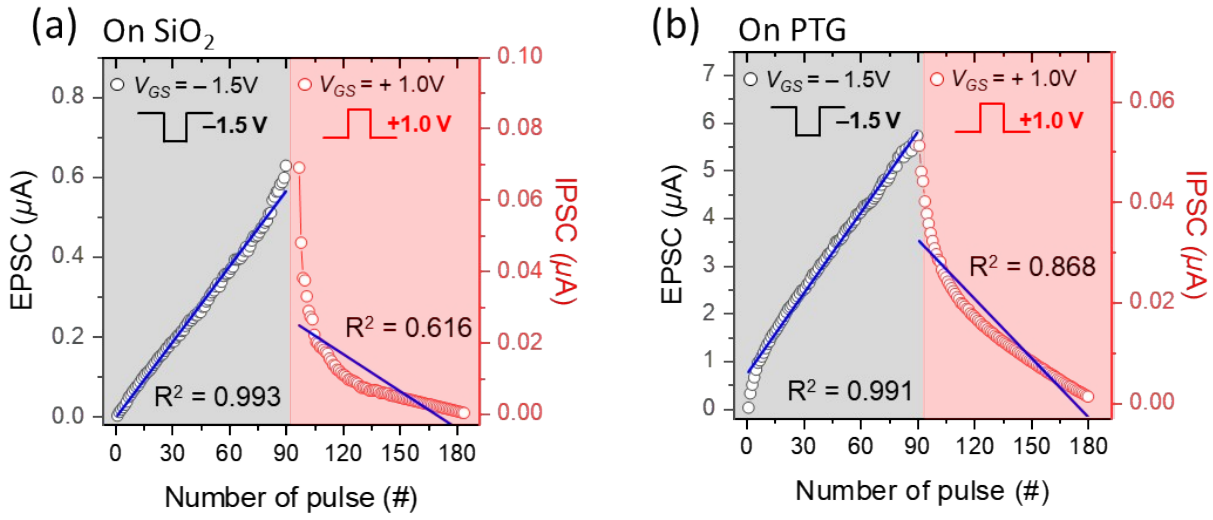

Figure S30. LTP/LTD characteristics of PBTTT OECT formed (a) on SiO_2 and (b) PTG film under the application of 90 potentiation pulses at –1.5 V and 87 depression pulses at +1.0 V.