Electronic Supplementary Information

Enhanced Hydrolytic and Electrical Stability of Eco-Friendly-Processed Polyimide Gate Dielectrics for Organic Transistors

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S2



Fig. S2 FTIR spectra of O-PAA annealed in the temperature range from 40 $^{\circ}$ C to 350 $^{\circ}$ C.



Fig. S3 FTIR spectra of W-PAAS annealed in the temperature range from 40 $^\circ$ C to 350 $^\circ$ C.



Fig. S4 (a,b) Optical microscope and (c,d) 3D confocal laser optical microscope images of O-PI thin

films prepared under humid air. The scale bar represents 50 $\mu m.$



Fig. S5 AFM images of spin-coated (a) O-PI prepared under ambient, (b) O-PI prepared under

inert, (c) W-PI prepared under ambient, and (d) W-PI prepared under inert.



under ambient, (b,f) O-PI prepared under inert, (c,g) W-PI prepared under ambient, and (d,h) W-

PI prepared under inert.

Table S1 Contact angle of DI water and diiodomethane droplets deposited on O-PI and W-PI thinfilms.

| Air condition | Contact angle [°] | | | |
|---------------|---|---|--|--|
| | DI water | Dioodomethane | | |
| ambient | 63.69 | 16.79 | | |
| inert | 64.39 | 17.14 | | |
| ambient | 58.06 | 13.31 | | |
| inert | 57.40 | 14.74 | | |
| | Air condition _ ambient inert ambient inert | Air condition Conta DI water ambient 63.69 inert 64.39 ambient 58.06 inert 57.40 | | |



inert, (c) W-PI prepared under ambient, and (d) W-PI prepared under inert before (top row) and after (bottom row) breakdown phenomena. The scale bars represent 200 μm.

| Table S2 N | Material and | dielectric p | roperties o | f O-PI and | W-PI prepa | red under a | ambient an | d inert |
|------------|--------------|--------------|-------------|------------|------------|-------------|------------|---------|
| air | | | | | | | | |
| dir. | | | | | | | | |

| PI | Air | Film thickness | Surface | Surface | Dielectric | Dielectric | FWHM | |
|----------------|-----------|------------------------|-----------|-----------------------|-----------------------------------|-----------------------------------|-------------------|----------|
| type condition | condition | Reduction ^a | roughness | energy | constant | loss at 100 Hz | r گ–11 | [nm] |
| | condition | [%] | [Å] | [mJ m ⁻²] | at 100 Hz | [tanδ, 10 ⁻³] | [A ⁻] | [1111] |
| O-PI | Ambient | 29.8 | 6.22 | 52.71 | $\textbf{3.28}\pm\textbf{0.08}$ | $\textbf{3.10}\pm\textbf{0.26}$ | 0.055 | 10.4 |
| | inert | 60.1 | 5.09 | 52.51 | $\textbf{3.23}\pm\textbf{0.06}$ | $\textbf{2.85} \pm \textbf{0.30}$ | 0.057 | 10 |
| W-PI | Ambient | 100 | 4.23 | 55.37 | $\textbf{3.26} \pm \textbf{0.05}$ | $\textbf{3.00} \pm \textbf{0.17}$ | 0.113 | 5 |
| | inert | 100 | 3.97 | 55.34 | $\textbf{3.18}\pm\textbf{0.03}$ | $\textbf{3.24}\pm\textbf{0.30}$ | 0.106 | 5.4 |

^{a)} Film thickness reduction estimated after 120 min exposure time.

^{b)} Dielectric properties are averaged from over 10 devices.



Fig. S8 2D GIXD patterns of (a) O-PI and (b) W-PI thin films prepared under inert. (c) Out-of-plane

and (d) in-plane 2D GIXD patterns of O-PI and W-PI thin films.



Fig. S9 Cross cutting test results for (a) O-PI and (b) W-PI thin films. The scale bar represents 20

μm.



Fig. S10 Transfer curves for C_{10} -BTBT-based OFETs with (a) O-PI in ambient, (b) O-PI in inert, (c) W-PI in ambient, and (d) W-PI in inert gate dielectrics. Blue and red lines represent I_{DS} and I_{GS} , respectively. Each figure contains 16 transfer curves measured from the same device sample.



20 [°] Fig. S11 Out-of-plane XRD patterns for C_{10} -BTBT thin films deposited on O-PI prepared under ambient, O-PI prepared under inert, W-PI prepared under ambient, and W-PI prepared under inert.



Fig. S12 AFM images of 40 nm thick C₁₀-BTBT thin films deposited on (a) O-PI prepared under ambient, (b) O-PI prepared under inert, (c) W-PI prepared under ambient, and (d) W-PI prepared under inert.



Fig. S13 AFM images of 5 nm thick C₁₀-BTBT thin films deposited on (a) O-PI prepared under ambient, (b) O-PI prepared under inert, (c) W-PI prepared under ambient, and (d) W-PI prepared under inert.



Fig. S14 Typical transfer curves for OFETs based on pentacene with (a) O-PI prepared under ambient, (b) O-PI prepared under inert, (c) W-PI prepared under ambient, and (d) W-PI prepared under inert. The black solid, black dash, and blue solid lines represent I_{DS} , I_{GS} , and $I_{DS}^{0.5}$, respectively.



Fig. S15 Out-of-plane XRD patterns for pentacene thin films deposited on O-PI prepared under ambient, O-PI prepared under inert, W-PI prepared under ambient, and W-PI prepared under inert.

Table S3 Electrical characteristics of OFETs based on pentacene with O-PI and W-PI as gatedielectrics prepared under ambient and inert air.

| PI type | Air condition | $\mu_{\text{FET}} [\text{cm}^2 \cdot \text{V}^{-1} \cdot \text{s}^{-1}]$ | <i>V</i> _{TH} [V] | $I_{\rm on}/I_{\rm off} [imes 10^4]$ | Yield [%] |
|---------|---------------|---|-------------------------------------|---|-----------|
| O-PI | ambient | 0.07 ± 0.02 | $\textbf{-9.57}\pm0.79$ | $\textbf{0.99} \pm \textbf{0.04}$ | 50 |
| | inert | $\textbf{0.05}\pm\textbf{0.01}$ | $\textbf{-10.59} \pm \textbf{0.67}$ | 1.50 ± 1.05 | 75 |
| W-PI | ambient | $\textbf{0.11}\pm\textbf{0.02}$ | $\textbf{-10.67} \pm \textbf{1.26}$ | $\textbf{4.22} \pm \textbf{1.78}$ | 100 |
| | inert | $\textbf{0.14}\pm\textbf{0.03}$ | $\textbf{-10.18} \pm 0.67$ | $\textbf{7.85} \pm \textbf{3.05}$ | 100 |

* Averaged over 16 devices.