Selective Reaction at Grain Boundaries Addressing Organic Field Effect Transistor Trap States

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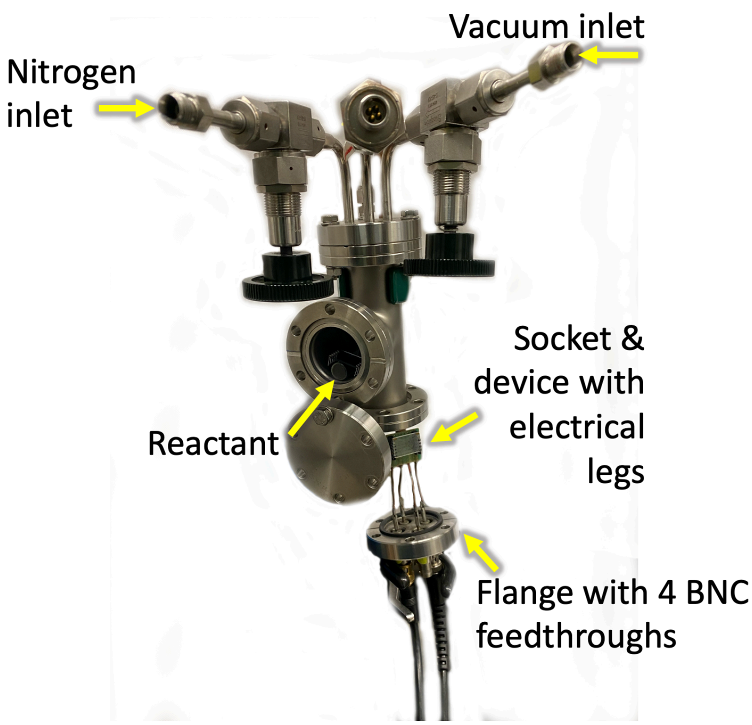

***Figure S1**. Reaction chamber for source-drain measurements and description of use.*

The reaction chamber was an ultra-high vacuum (UHV) Tee made of stainless steel. One end of the Tee was used to house the reactant, typically in an open glass via just inside the metal tube, and was sealed with a UHV flange during reaction. The lower end of the Tee had a modified flange that consisted of BNC feedthroughs that led to a socket holder. The various BNC feedthroughs led to different sources and a common drain. Samples that were not monitored in real time were placed in the base of this flange when the system was sealed. The topmost end of the Tee terminates a flange modified with two VCR bellows valves and a mount for a pressure gauge. This flange is left in place, though the two VCR connections are hooked up to a vacuum line and nitrogen line allowing the atmosphere of the chamber to be controlled.

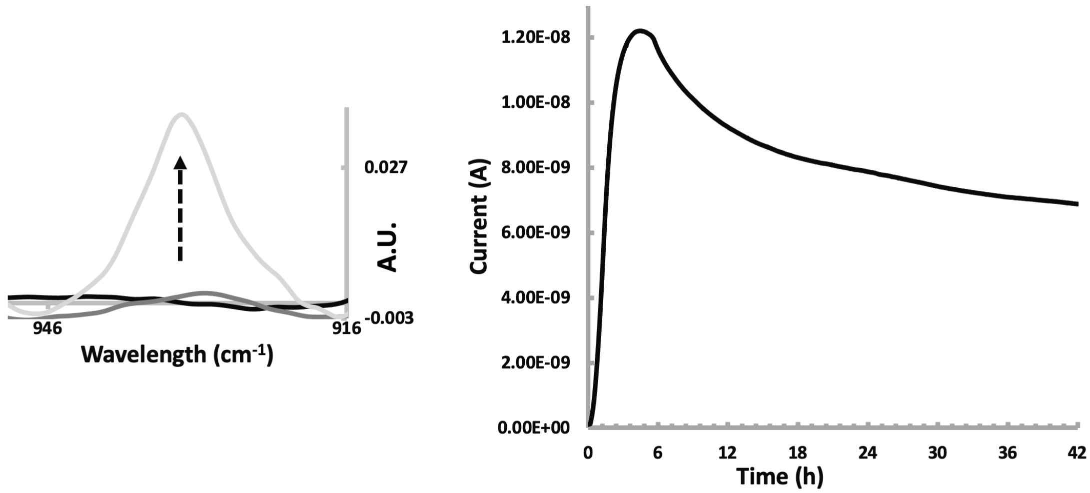

Figure S2*. Additional Spectroscopic Reactivity Data Corroborated with Conductance Changes.* Left: Additional regions of the witness sample described in Fig 2. Right: Real-time change in conductance for the sample characterized by UV-vis in Fig 2. The large increase in conductance at the beginning eventually begins to decrease, presumed to be a result of the consumption of the active channel, or reorientation/randomization of the reaction product reducing the dipole's effect on the mean energy within grains.

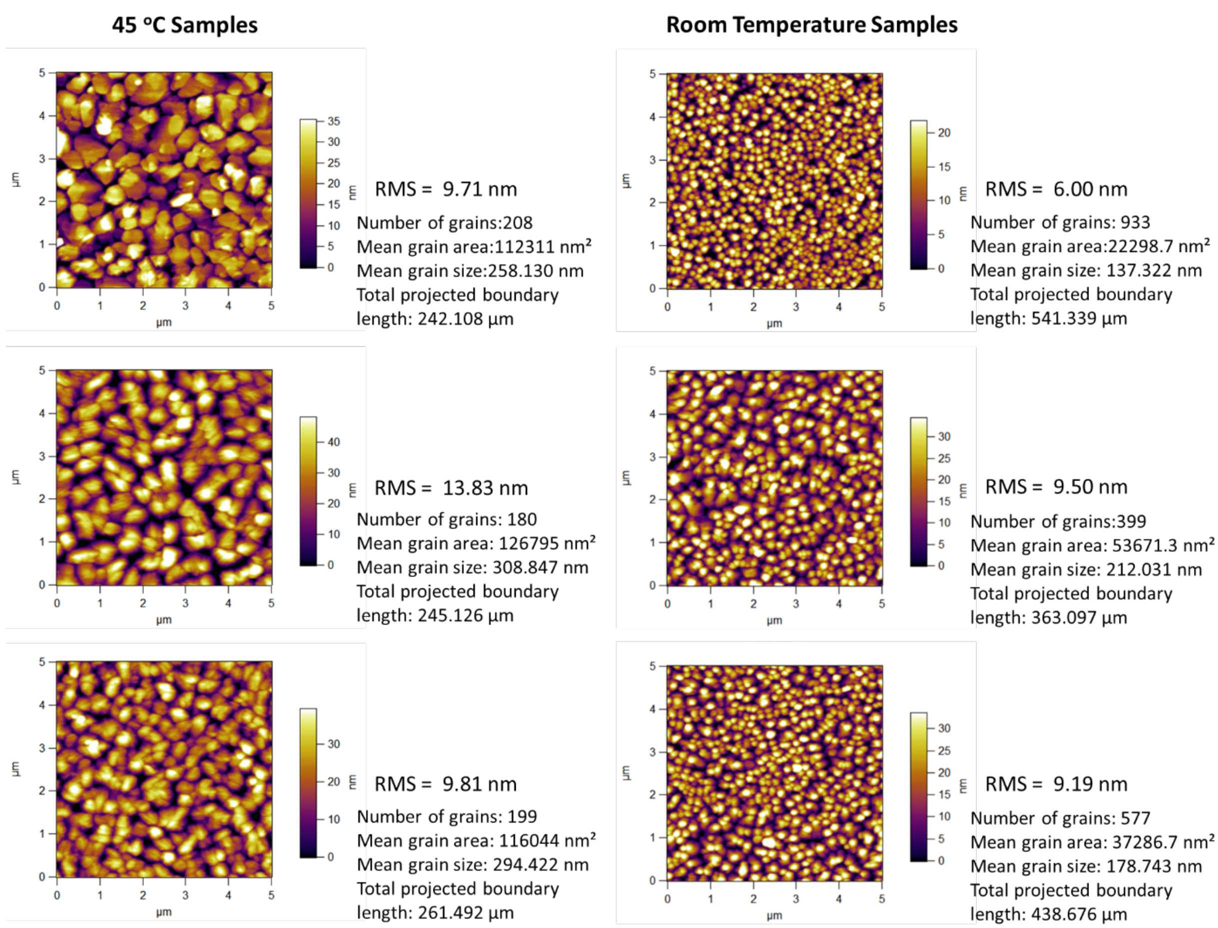

Figure S3*. AFM measurements of grain size and average grain boundary length*

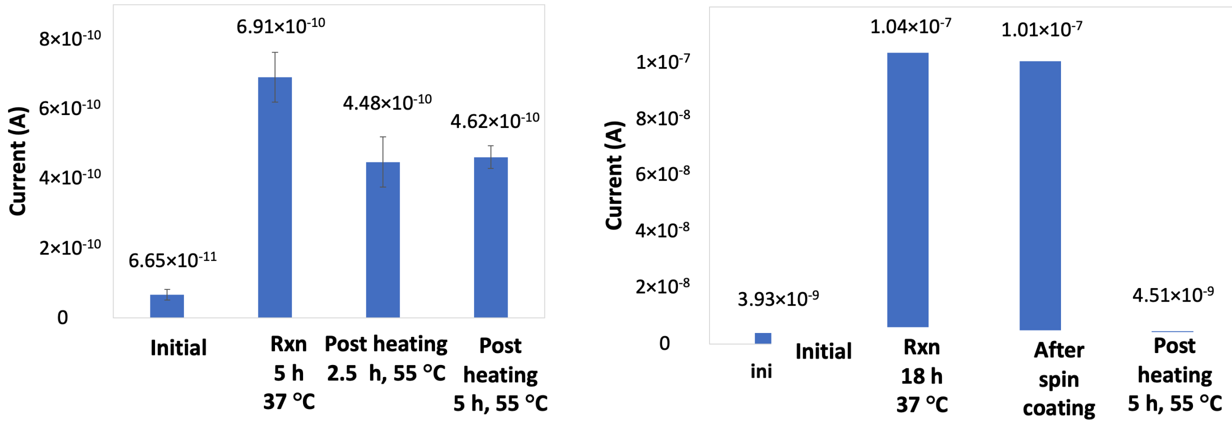

Figure S4. *Device Stability Under Exposure to CYTOP.*

Conductivity of pentacene (100 nm) between two bottom-contact gold electrodes at 5 V, exposed to various conditions. The left graph is for samples that were reacted and then heated without CYTOP. The right graph shows a similar sample that was exposed to the CYTOP formulation after reaction, and then later heated.

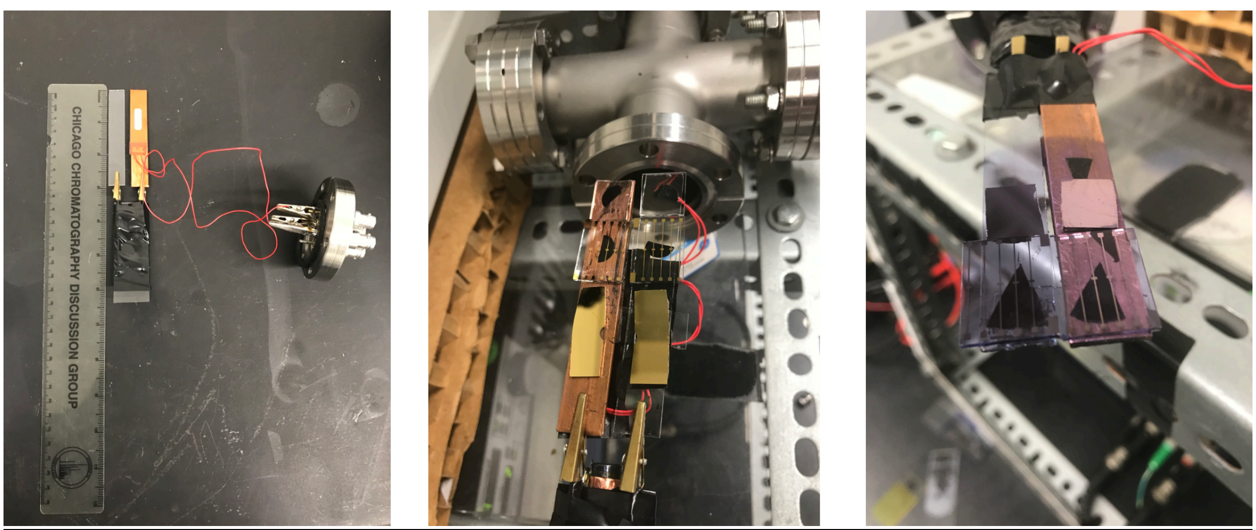

Figure S5. *Photos of Setup to Control Substrate Temperature During Pentacene Deposition.*

For the room temperature samples, a microscope slide was cut to length (7.5 cm × 1.2 cm) and devices were attached with carbon tape. For heating the substrate, a polyimide flexible resistive heater (Omega) was attached to a copper bar (7.5 cm × 1.2 cm) and was attached approximately 1 cm away from the glass substrate. The substrates were attached to the copper bar via carbon tape at the edges of the samples. The leads for the heater led out of the sublimation chamber to via BNC feedthrough in the flange that led to a power supply. The voltage was set to 10 V which heated the sample to approximately 45 °C after 20 minutes.

Bottom Gate CYTOP OFET Measurements

Bottom contact, bottom gate OFETs with CYTOP dielectric were prepared by first depositing Ti/Au contacts onto a Kapton substrate as a gate electrode. Next, a layer of CYTOP was spin-coated onto the substrate at 2000 RPM with a ramp of 2 s and a hold of 60 s. The substrate was then placed on a hotplate at 110 °C for 25 min to cure the CYTOP. Source and drain electrodes were deposited through a shadow mask and consisted of 40 nm Au, similar to the other OFETs included in this study. Finally, the pentacene film (100 nm) was thermally evaporated over these predefined structures.

The areal capacitance of the dielectric was measured using an Agilent LCR meter and an average value of 2.3 $\times 10^{-8}$ F/cm^2 was obtained.

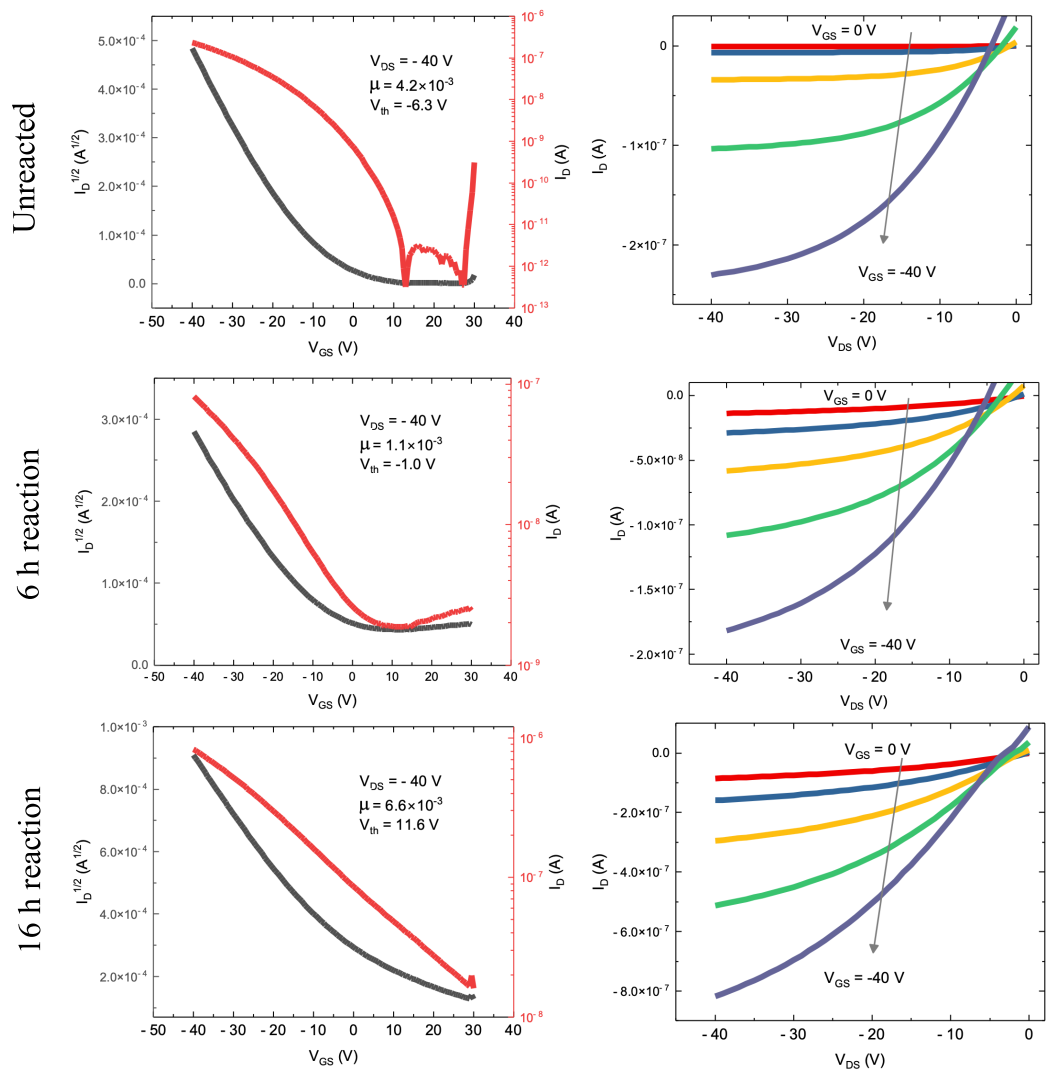

Figure S6. *I-V measurements on Bottom Gate CYTOP OFETs.*

Representative transfer (left) and output characteristics (right) for OFET in a bottom contact, bottom gate configuration with CYTOP dielectric and 100 nm of pentacene. From top to bottom: unreacted pentacene, pentacene that has been reacted at 37 °C for 6 h, pentacene that has been reacted at 37 °C for 16 h.