Supporting Information for:

Operating and Sensing Mechanism of Electrolyte-Gated Transistors with Floating Gates: Building a Platform for Amplified Biodetection

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1. Additional images of the device architecture. Figure S1 depicts an image of a printed P3HT film, an optical image of an FG-EGT, and a schematic of the microfluidic channels used to selectively functionalize the floating gate with SAMs.

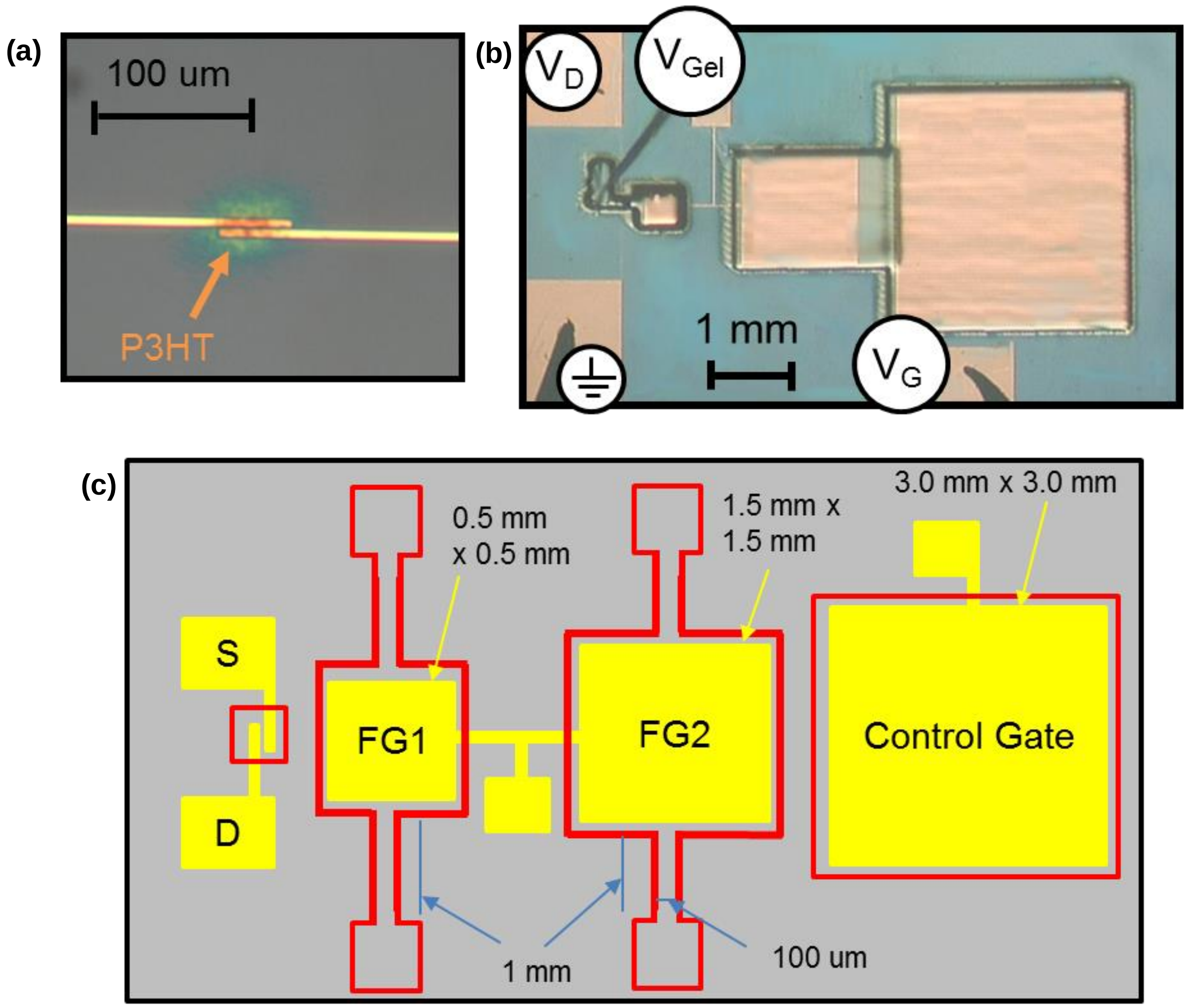

Figure S1: Device Structure. (a) is an image of the printed P3HT film (50 nm) with W/L = 40 μm/2 μm. **(b)** is an image of an FG-EGT with $A_{FG1} = 150A_{P3HT}$, $A_{FG2} = 1500A_{P3HT}$ and $A_{CG} = 6000A_{P3HT}$. **(c)** is a schematic of the FG-EGT with the microfluidic channels (outlined in red) used to selectively functionalize the floating gate with SAMs.

2. Derivation of Equations 2-3

Considering the circuit in Figure 1C (two capacitors in series), a charge balance yields

$$Q_{Gate} + Q_{P3HT} = 0 \tag{S1}$$

substituting in the definition of capacitance (Q = CV)

$$C_{Gate}(V_{Gel} - V_G) + C_{P3HT} V_{Gel} = 0 \tag{S2}$$

and rearranging for Equation 2 after substituting $C = C_i A$

$$V_{Gel} = \frac{1}{1 + \frac{C_{i,P3HT} A_{P3HT}}{C_{i,Gate} A_{Gate}}} V_G \tag{2}$$

Rearranging further results in Equation 3, the fraction of voltage dropped at the gate electrode

$$F^{Gate} = \frac{V_G - V_{Gel}}{V_G} = \frac{1}{1 + \frac{C_{i,Gate} A_{Gate}}{C_{i,P3HT} A_{P3HT}}} \tag{3}$$

To overcome experimental limitations, mainly the variability of open circuit voltage of the quasi-reference Pt electrode, we considered the slope of V_{Gel}-V_G

$$\frac{dV_G}{dV_{Gel}} = 1 + \frac{C_{i,P3HT} A_{P3HT}}{C_{i,Gate} A_{Gate}} \tag{S3}$$

When plotting this relationship, we found that the slope of dV_G/dV_{Gel} with respect to A_{P3HT}/A_{Gate} is ~8, representing the ratio of specific capacitances. However, the intercept was not unity, and instead 1.17. We attribute this area-independent potential drop to a combination of series resistance through the ion-gel bulk and a parallel resistance at the gate electrode interface due to charge transfer and electrode polarization. This is sketched in Figure S2.

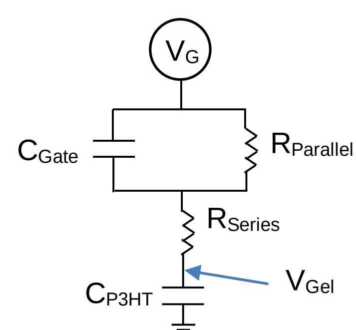

Figure S2: Sketch of a more advanced equivalent circuit of an EGT.

3. Additional data on hysteresis effects in FG-EGTs. In addition to altering the relationship between V_G and V_{Gel}, lowering the gate area also affects the hysteresis in an EGT. We observed a systematic increase in hysteresis, defined as the difference in V_G between the forward and reverse sweep at the same I_D (Figure S3A), with lowered gate area (Figure S3B). We attributed this to a combination of potential dependent capacitance, overpotentials associated with interfacial charge transfer, and mass transport limitations of ions into the P3HT film as discussed in the main text.

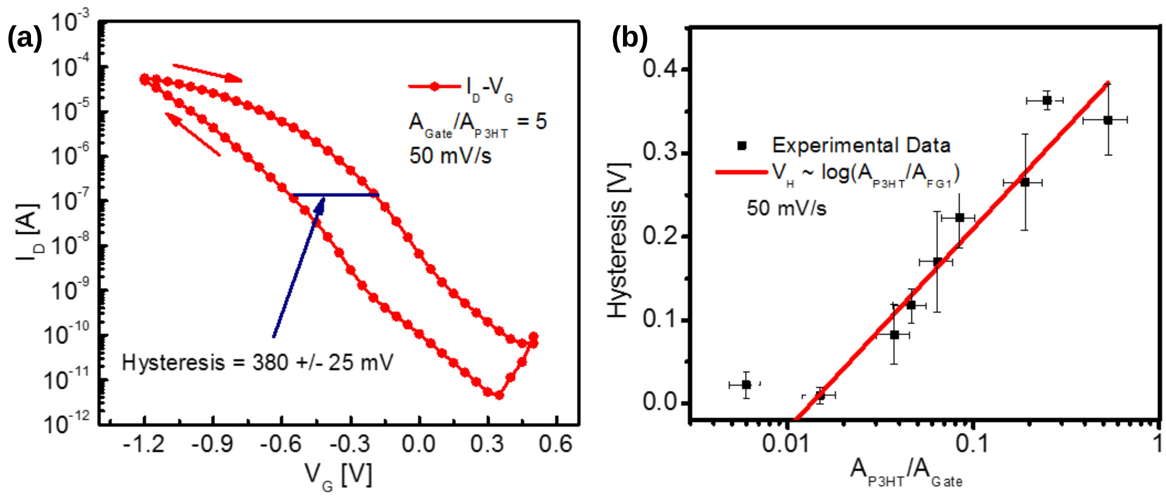

Figure S3: Hysteresis in an EGT. (a) is an EGT with $A_{Gate}/A_{P3HT} = 5$ and a significant hysteresis of 380 ± 25 mV, calculated by measuring the difference in V_G between the forward and reverse sweep at the same I_D. This calculation is repeated for multiple A_{Gate} values and plotted in (b) with a logarithmic fit in red.

4. Derivation of Equations 4-6

Considering the circuit in Figure 1F (two capacitors in series and an additional parasitic capacitor), we compared the effect of applying V_G to the floating gate (V_G^{EGT}) and control gate (V_G^{CG}) by performing a charge balance and assuming I_D directly reflects the charge on C_{1E} (Q_{1E}).

$$\Delta Q_{1E} = 0 \tag{S4}$$

Substituting in the definition of capacitance (Q = CV) yields

$$C_{1E}V_G^{EGT} - C_{1E}V_F = 0 \tag{S5}$$

Next, perform a charge balance on the node labeled "V_F" in Figure 1F while assuming Q* = 0 to express V_F in terms of V_G^{CG} analogous to the derivation of Equation 2

$$V_F = \frac{1}{1 + \frac{C_{1E}}{C_{2E}}} V_G^{CG} \tag{S6}$$

Substitute Equation S6 into Equation S5 for Equation 4 in the text,

$$V_G^{CG} = \left(\frac{C_{1E}}{C_{2E}} + 1\right) V_G^{EGT} \tag{4}$$

Next, introducing a stray capacitor with Q* stored on it requires modification of the charge balance on the node labeled "V_F"

$$Q_{1E} + Q_{2E} + Q^* = 0 \tag{S7}$$

Substituting in the definition of capacitance furnishes

$$C_{1E}V_F + C_{2E}(V_F - V_G^{CG}) + Q^* = 0 \tag{S8}$$

Rearranging results in Equation 5 in the text,

$$V_F = \frac{C_{2E}}{C_{1E} + C_{2E}} V_G^{CG} - \frac{Q^*}{C_{1E} + C_{2E}} \tag{5}$$

To account for Q* empirically, we assumed that it is a constant fraction of the charge stored in C_{2E}, but opposite in sign, so that $f = Q^*/-Q_{2E}$ and substituted this into Equation 5

$$V_F = \frac{C_{2E}}{C_{1E} + C_{2E}} V_G^{CG} + \frac{f C_{2E}(V_F - V_G^{CG})}{C_{1E} + C_{2E}} \tag{S9}$$

Rearranging for V_F in terms of V_G^{CG} yields

$$V_F = (1 - f)\frac{1}{1 + \frac{C_{1E}}{C_{2E}}} V_G^{CG} + f \frac{1}{1 + \frac{C_{1E}}{C_{2E}}} V_F \tag{S10}$$

$$\left((1 - f) + \frac{C_{1E}}{C_{2E}} \right) V_F = (1 - f) V_G^{CG} \tag{S11}$$

$$V_F = \frac{1}{1 + \frac{1}{1 - f}\frac{C_{1E}}{C_{2E}}} V_G^{CG} \tag{S12}$$

Substituting Equation S12 back into Equation S5 above results in Equation 6 in the text,

$$V_G^{CG} = \left(\frac{C_{1E}}{C_{2E}} \frac{1}{1 - f} + 1 \right) V_G^{EGT} \tag{6}$$