

## SUPPORTING INFORMATION

# Poly(3-hexylthiophene)-based Organic Thin-Film Transistors with Virgin Graphene Oxide as an Interfacial Layer

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### Calculation of Surface and Interfacial Energies

In the Owens-Wendt model, the surface energy is divided into two components: a polar component and a dispersive component of non-polar interaction. Therefore, the surface energy is given by:<sup>1-2</sup>

$$\gamma_S = \gamma_S^d + \gamma_S^P \quad (\text{S1})$$

where  $\gamma_S$  is the surface free energy of a solid;  $\gamma_S^d$  and  $\gamma_S^P$  are the dispersive and polar components of the surface free energy of a solid, respectively. The dispersive and polar components of the surface free energy of a solid can be determined using the following equation:<sup>3-4</sup>

$$\gamma_L(1 + \cos\theta) = 2\sqrt{\gamma_S^d \gamma_L^d} + 2\sqrt{\gamma_S^P \gamma_L^P} \quad (\text{S2})$$

where  $\gamma_L$  is the surface free energy of an immersion liquid,  $\gamma_L^d$  and  $\gamma_L^P$  are the surface free energy of the dispersive and polar components of an immersion liquid, respectively, and  $\theta$  is the contact angle of a liquid on the solid surface. In Equation **S2**, since the values for  $\gamma_L^d$ ,  $\gamma_L^P$  and  $\gamma_L$  (where  $\gamma_L = \gamma_L^d + \gamma_L^P$ ) are known<sup>5</sup>, and  $\theta$  can be measured, we have only two unknowns,  $\gamma_S^d$

and  $\gamma_S^P$ ; therefore, we need two contact angles from two different liquids to solve for the two unknowns. **Eq. S2** can be re-written for the two liquids as follows:

$$\gamma_{L1}(1 + \cos\theta_1) = 2\sqrt{\gamma_S^d \gamma_{L1}^d} + 2\sqrt{\gamma_S^P \gamma_{L1}^P} \quad (\text{S3})$$

$$\gamma_{L2}(1 + \cos\theta_2) = 2\sqrt{\gamma_S^d \gamma_{L2}^d} + 2\sqrt{\gamma_S^P \gamma_{L2}^P} \quad (\text{S4})$$

The two liquids used to measure the contact angles on the SiO<sub>2</sub> and GO surfaces are hexadecane (C<sub>16</sub>H<sub>34</sub>) and water. Hexadecane was chosen for its non-polar nature, and water was chosen for its polar nature. The standard deviation for the contact angle measurements was  $\pm 2$  degrees. The surface free energy of the dispersive and polar components of hexadecane and water are shown in **Table S1**.

Table S1: Surface free energy of the dispersive and polar component of hexadecane and water  
(taken from Ref. 5)

	<b>Water</b>	<b>Hexadecane</b>
$\gamma_L^d$ (mN/m)	21.8	26.35
$\gamma_L^P$ (mN/m)	51	0
$\gamma_L$ (mN/m)	72.8	26.35

The contact angles of water on the SiO<sub>2</sub> and GO surfaces are 28° and < 5°, respectively. The contact angles of hexadecane on SiO<sub>2</sub> and GO surfaces are < 5°. The dispersive and polar components of the surface free energy of SiO<sub>2</sub> and GO are determined using **Eqs. S3** and **S4**, and the results are shown in **Table S2**. The surface free energy ( $\gamma_S$ ) of SiO<sub>2</sub> and GO are determined

using **Eq. S1**, and the results are also shown in **Table S2**. Surface energy and its components for P3HT (available from the scientific literature)<sup>6</sup> are also presented in **Table S2**.

Table S2: Surface free energy of SiO<sub>2</sub>, GO, and P3HT.

	SiO <sub>2</sub>	GO	P3HT
$\gamma_S^d$ (mN/m)	26.35	26.4	24.6
$\gamma_S^p$ (mN/m)	38.9	46.8	2.3
$\gamma_S$ (mN/m)	65.25	73.2	26.9

After obtaining the surface energies of all the materials (**Table S2**), the interfacial energy between two materials can be calculated using the following equation:<sup>1</sup>

$$\gamma_{1-2} = \gamma_1 + \gamma_2 - \frac{4\gamma_1^d \gamma_2^d}{\gamma_1^d + \gamma_2^d} - \frac{4\gamma_1^p \gamma_2^p}{\gamma_1^p + \gamma_2^p} \quad (\text{S5})$$

The interfacial energy of P3HT/SiO<sub>2</sub> and P3HT/GO interfaces are determined using **Eq. S5**, and the results are shown in **Table S3**.

Table S3: Interfacial energy of P3HT/SiO<sub>2</sub> and P3HT/GO

	P3HT/SiO <sub>2</sub>	P3HT/GO
$\gamma_{1-2}$ (mN/m)	32.58	40.39

## References

1. Wu, S., Calculation of interfacial tension in polymer systems. *Journal of Polymer Science Part C: Polymer Symposia* **1971**, 34 (1), 19-30.

2. Luzinov, I.; Xi, K.; Pagnouille, C.; Huynh-Ba, G.; Jérôme, R., Composition Effect on the Core–Shell Morphology and Mechanical Properties of Ternary Polystyrene/Styrene–Butadiene Rubber/Polyethylene Blends. *Polymer* **1999**, *40* (10), 2511-2520.
3. Owens, D. K.; Wendt, R. C., Estimation of the surface free energy of polymers. *J. Appl. Polym. Sci.* **1969**, *13* (8), 1741-1747.
4. Żenkiewicz, M., Methods for the calculation of surface free energy of solids. *Journal of achievements in materials and manufacturing engineering* **2007**, *24*, 137-145.
5. Janczuk, B.; Wojcik, W.; Zdziennicka, A.; Bruque, J. M., Components of the surface free energy of low rank coals in the presence of n-alkanes. *Powder Technology* **1996**, *86* (3), 229- 238.
6. Wang, X.; Ederth, T.; Inganäs, O., In Situ Wilhelmy Balance Surface Energy Determination of Poly(3-hexylthiophene) and Poly(3,4-ethylenedioxythiophene) during Electrochemical Doping–Dedoping. *Langmuir* **2006**, *22* (22), 9287-9294.