Electronic Supplementary Information

Comparing different geometries for photovoltaicthermoelectric hybrid devices based on organics

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Appendix A

The overall output power from a hybrid PV-TE device, $P_{H,out}$, is given by:

$$P_{H,out} = P_{PV} + P_{TEG}$$
(A.1)
where P_{PV} is output power from the solar cell and P_{TEG} is output power from the TEG.

In terms of efficiency, Equation 1 can be rewritten as

 $\eta_H(T) = \eta_{PV}(T) + \eta_{TEG}(T)$ (A.2) where η_H is the overall efficiency of the hybrid device, η_{PV} is the solar cell efficiency, and η_{TEG} is the efficiency of the TEG. Solar cell light-to-electricity efficiency is dependent on several factors, such as temperature (*T*), reflectance, transmittance, charge carrier separation efficiency, charge carrier collection efficiency, etc. To emphasize that the temperature of the PV system can increase when coupling a TEG, we make the temperature dependence in Equation A.2 explicit.

For the **<u>non-contact geometries</u>**, it should be noted that $\eta_{TEG} = \eta_{SOTEG}$, where η_{SOTEG} is the efficiency of the SOTEG. Following Chen et. al. [41], η_{SOTEG} can be defined as

 $\eta_{SOTEG} = \eta_{opt}\eta_{abs}\eta_{teg}\eta_{aux}$ (A.3) where η_{opt} is the optical efficiency, η_{abs} is the absorber efficiency, η_{teg} is the efficiency of a thermoelectric generator, and η_{aux} is the auxiliary efficiency. The auxiliary efficiency accounts for any parasitic system losses, such as electricity consumption for active cooling. The thermal losses arising from convection and radiation losses, for example, are included in the TEG efficiency. Assuming no optical concentration and no auxiliary device losses, Equation A.3 can be reduced to

 $\eta_{SOTEG} = \eta_{abs} \eta_{teg}$ (A.4) For devices with non-contact modes (reflection and transmission geometries), the heat generated by the solar cell will not contribute to the SOTEG efficiency, as a gap is left between the solar cell and the SOTEG. Only transmitted or reflected light from the solar cell that is absorbed by the TEG can be converted into electricity, and the very small emissive component of the solar cell is neglected. Now, for the reflection geometry, we consider an opaque cell and neglect the small emissivity of the cell at room temperature. In this case, the hybrid device efficiency becomes:

$$\eta_{H}^{Reflection\,geometry} = \eta_{PV} + R \eta_{teg} \eta_{abs} \tag{A.5}$$

where *R* is the reflectance from the PV cell. For the transmission and non-contact geometry:

$$\eta_{H}^{Transmission \ geometry} = \eta_{PV} + \tau \eta_{teg} \eta_{abs}$$
(A.6)

where τ is the transmittance from a PV cell.

For the case of hybrid devices with a **<u>contact geometry</u>**, the SOTEG converts thermal and optical energy into electricity, thus the SOTEG efficiency can be defined as

$$\eta_{TEG}(T) = \eta_{TEG}^{opt} + \eta_{TEG}^{heat}$$
(A.7)

In contact mode, one must consider a solar cell's light-to-heat, or photothermal, efficiency as the heat generated by the solar cell contributes to the energy the SOTEG can convert into electricity. The photothermal efficiency of a solar cell, $\eta_{PV,PT}$, can be given as

$$\eta_{PV,PT} = (1 - \eta_{PV})\eta_{opt,PV} \tag{A.8}$$

where $\eta_{opt,PV}$ is the optical efficiency of the solar cell, which accounts for the sum of reflectance, transmittance and emission losses, i.e. $\eta_{opt,PV} = R + \tau + E$.

For the case of contact mode with opaque cells, $\eta_{TEG}^{opt} = 0$ because there is no transmitted light:

$$\eta_{H}^{Contact opaque} = \eta_{PV} + (1 - \eta_{PV})\eta_{opt, PV}\eta_{H, coupling}\eta_{teg}$$
(A.9)

where $\eta_{H,coupling}$ is a coupling term to describe the SOTEG's ability to make use of the heat generated by the solar cell.

For a device with a contact geometry and semitransparent cells, the device efficiency can be written as:

$$\eta_{H}^{Contact \ semitransparent}(T) = \eta_{PV}(T) + \eta_{teg} (\eta_{TEG,PT} \tau + (1 - \eta_{PV}) \eta_{opt,P}$$
(A.10)

Temperature dependence of efficiency for a Silicon solar cell



Figure S1- Silicon solar cell efficiency and temperature under continuous illumination of 1 Sun.

In Figure S1, the efficiency of the commercial silicon solar cells displays a clear negative temperature coefficient, with efficiency decreasing by more than 1.5 percentage points.

Temperature evolution of PCE11 and other PV systems with different support materials



Figure S2- a) Temperature evolution of PCE11 with different support materials; b) Steady-state temperature achieved by the PCE11 vs support materials and c) Temperatures of different OPV materials in contact with support materials.

To investigate the role of support materials on steady-state temperature achieved under illumination, we measured the temperature of a free-standing film of PCE11 under illumination in contact with different support materials: a glass substrate, a PET substrate, and a slab of black PLA. In Figure S2a, placing a support material allowed the free-standing film to achieve higher temperatures, with the highest temperature being achieved with the black PLA slab. Black PLA has a low-thermal conductivity and absorbs light transmitted by the OPV material, thus allowing the material to reach higher temperatures [27]. Additionally, the black PLA serves as a barrier to convection, further enhancing temperature. The temporal evolution of temperature for PCE11 in contact with black PLA is much slower because of the added thermal mass of the black PLA slab. The final steady state temperatures are shown in Figure S3c.

In Figure S2a, different OPV materials were placed in contact with a PET substrate and the same black PLA slab. We observed that in all cases, the materials achieved the highest temperature when in contact with black PLA. The majority of the temperatures are very close together, lying in a range between 323 and 327 K, which likely is a result of the additional absorption from the PLA.



Figure S3 - Schematic of the photothermal experiment.

The emissivities used for the organic materials were approximately 0.90.



Figure S4 – Specular reflectance data for OPV materials.



Figure S5 – Specular reflectance data for active layer OPV materials.



Figure S6 – Transmittance data for OPV materials.



Figure S7 – Transmittance data for the active layer OPV materials.

The cut-on wavelength is defined as the point where transmittance exceeds 10% (dashed red lines). For example, in the case of P3HT, the cut-on wavelength is 660 nm.

Measured power densities

Material	Transmitted	Reflected
	Power	Power
	mW cm ⁻²	mW cm ⁻²
Air	90.00	-
Glass	80.90	20.24
ZnO	85.61	22.92
ΙΤΟ	67.04	27.12
ITOZnO	69.33	24.96
PFO	64.49	26.48
MEHPPV	61.75	21.01
PFBT	35.66	21.90
РЗНТ	39.27	18.33
PCE12	43.00	23.81
PCE13	36.25	17.32
TQ1	36.15	15.02
PCE10	41.35	17.06
PCE11	27.07	18.59
N2200	32.01	21.14
OPV	6.96	14.64
PLA (White)	-	14.90
SiCell	-	9.17
Mirror	-	69.14

Table S1 - Measured power densities

zT-dependent TEG Efficiencies



Figure S8 - a) Calculated TEG efficiencies for eDIPS:Cellulose for varying zTs in reflection mode: 0.5 (Black square), 0.01 (Wine Square), 0.1 (orange square), 1 (purple square), and 2.5 (blue square). The dashed black line represents the TEG efficiency calculated assuming a zT of 2.5 with no filter above (100% Reflection); b) calculated power densities for DMSO-treated PEDOT:PSS.



Figure S9 - a) Calculated TEG efficiencies for eDIPS:cellulose for varying zTs: 0.5 (Black square), 0.01 (Wine Square), 0.1 (orange square), 1 (purple square), and 2.5 (blue square) in non-contact transmission mode. The dashed black line represents the TEG efficiency calculated assuming a zT of 2.5 with no filter above (100% transmission); b) calculated power densities for DMSO-treated PEDOT:PSS.