Supplementary information

Determining the shunt resistance and photogenerated current of tandem organic solar cells via simulating their practical photovoltaic parameters

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1 Determining the average bandgap, energetic disorder and effective mobility of the single-junction sub-cells

1.1 The acquisition of average bandgap and energetic disorder of active layer

The active layers of the single-junction sub-cells approximate to the black bodies. Thus, the photogenerated current (J_{ph}) of each single-junction sub-cell obtained under 1 sun illumination can be estimated by

$$J_{ph} = aq\int_{E_{g,A}}^{\infty} \frac{AM1.5G}{E} dE \quad , \tag{S1}$$

where a is the absorptance of device, q is the elementary charge, AM 1.5G represents the spectral irradiance of 1 sun solar light, $E_{g,A}$ is the average bandgap of active layer, E is the photon energy. The experimentally measured J_{ph} is set to the reverse saturation current density of device experimentally measured under 1 sun, because the loss of short-circuit current density (J_{SC}) due to shunt resistance (R_{SH}) is retrieved at the reverse bias. The a is taken as the average experimentally measured external quantum efficiency (EQE) from 450 to the wavelength at the half height of the long-wavelength hypotenuse for the EQE spectrum. When the calculated J_{ph} based on Eq. (S1) is equal to the experimentally measured one under 1 sun illumination, the $E_{g,A}$ of active layer is concluded.

The open-circuit voltage (V_{OC}) of OSC is expressed as

$$V_{OC} = \frac{k_B T}{q} ln\left(\frac{J_{ph} - \frac{V_{OC}}{R_{SH}}}{J_0}\right) \quad , \tag{S2}$$

where k_B is Boltzmann constant, T is temperature, J_0 is the background current density due to the bimolecular recombination. Under 1 sun illumination, Eq. (S2) is approximated to

$$V_{OC} \approx \frac{k_B T}{q} ln\left(\frac{J_{SC}}{J_0}\right) \quad . \tag{S3}$$

Provided that the bimolecular recombination is predominant and the bandgap energies meet the Gaussian distribution in active layer, the J_0 is formulated by

$$J_0 = aJ_{00}exp(-\frac{E_{g,A}}{k_BT} + \frac{\sigma^2}{2k_B^2T^2}), \quad \text{(S4)}$$

where J_{00} is expressed as

$$J_{00} = \frac{2\pi q}{h^3c^2}(k_BTE_{g,A}^2 + 2k_B^2T^2E_{g,A} - 2\sigma^2E_{g,A} + \frac{\sigma^4}{k_BT} + 2k_B^3T^3 - k_BT\sigma^2), \quad \text{(S5)}$$

σ is energetic disorder equivalent to the standard deviation of the bandgap energies' Gaussian distribution, h is Planck constant, c is the speed of light. Based on the concluded $E_{g,A}$, experimentally measured J_{SC} and V_{OC} under 1 sun illumination, the σ of active layer is calculated via Eqs. (S3-5).

1.2 The calculation of illuminated *J-V* characteristics

The device current (J) under the illumination is calculated by

$$J = J_d + J_{SH} - J_{ph}, \quad \text{(S6)}$$

where J_d is the bimolecular recombination current, J_{SH} is the leakage current. The J_d is described by

$$J_d = J_0exp(\frac{qV_d}{k_BT}), \quad \text{(S7)}$$

where V_d is the quasi-Fermi levels' separation in active layer. The J_{SH} is formulated by

$$J_{SH} = \frac{V_d}{R_{SH}}. \quad \text{(S8)}$$

The applied voltage (V) of is calculated by

$$V = V_d + V_R, \quad \text{(S9)}$$

where V_R is the voltage drop resulting from the series resistance R_S. The V_R is expressed as

$$V_R = JR_S. \quad \text{(S10)}$$

The R_S is formulated as follows [S4,5]

$$R_S = \frac{d}{2q\mu_{eff}exp(\frac{qV_a}{2k_BT})\sqrt{N_HN_L}exp(\frac{-E_{g,A}}{2k_BT} + \frac{\sigma^2}{8k_B^2T^2})}, \quad \text{(S11)}$$

where d is active layer's thickness, μ_{eff} is effective carrier mobility defined as the square root of the product of hole mobility times electron mobility, N_H and N_L are the densities of states for highest occupied molecular orbital for hole hopping and lowest unoccupied molecular orbital for electron hoping in active layer, respectively.

1.3 The extractions procedure of μ_{eff} and R_{SH} via modeling the performance of single-junction sub-cell under 1 sun illumination

Firstly, the μ_{eff} and R_{SH} are preset to 1×10^{-4} cm^2 V^{-1} s^{-1} and 1000 Ω cm^2, respectively. Secondly, the V_d is preset from 0 to 2 V at step of 0.001 V. At each preset V_d, the J and V are calculated via Eqs. (S4-11), based on the deduced $E_{g,A}$, σ, measured J_{ph}, preset μ_{eff} and R_{SH}. By varying the preset values of μ_{eff} and R_{SH}, when the extracted J_{SC} and PCE from simulated *J-V* curve are identical to those measured for real device, the μ_{eff} and R_{SH} are concluded.

Note that, the drift–diffusion model is confirmed equivalent to the Shockley equation model in terms of simulating the *J-V* characteristics of organic solar cells.[S1,2]

References.

S1 U. Würfel, D. Neher, A. Spies and S. Albrecht, *Nat. Commun.*, 2015, **6**, 6951.

S2 A. Fischer, M. Pfalz, K. Vandewal, S. Lenk, M. Liero, A. Glitzky and S. Reineke, *Phys. Rev. Appl.*, 2018, **10**, 014023.