

# ***Analysis and optimization of organic tandem solar cells by full opto-electronic simulation – Supplementary Material***

## **1 OPTICAL MATERIAL DATA**

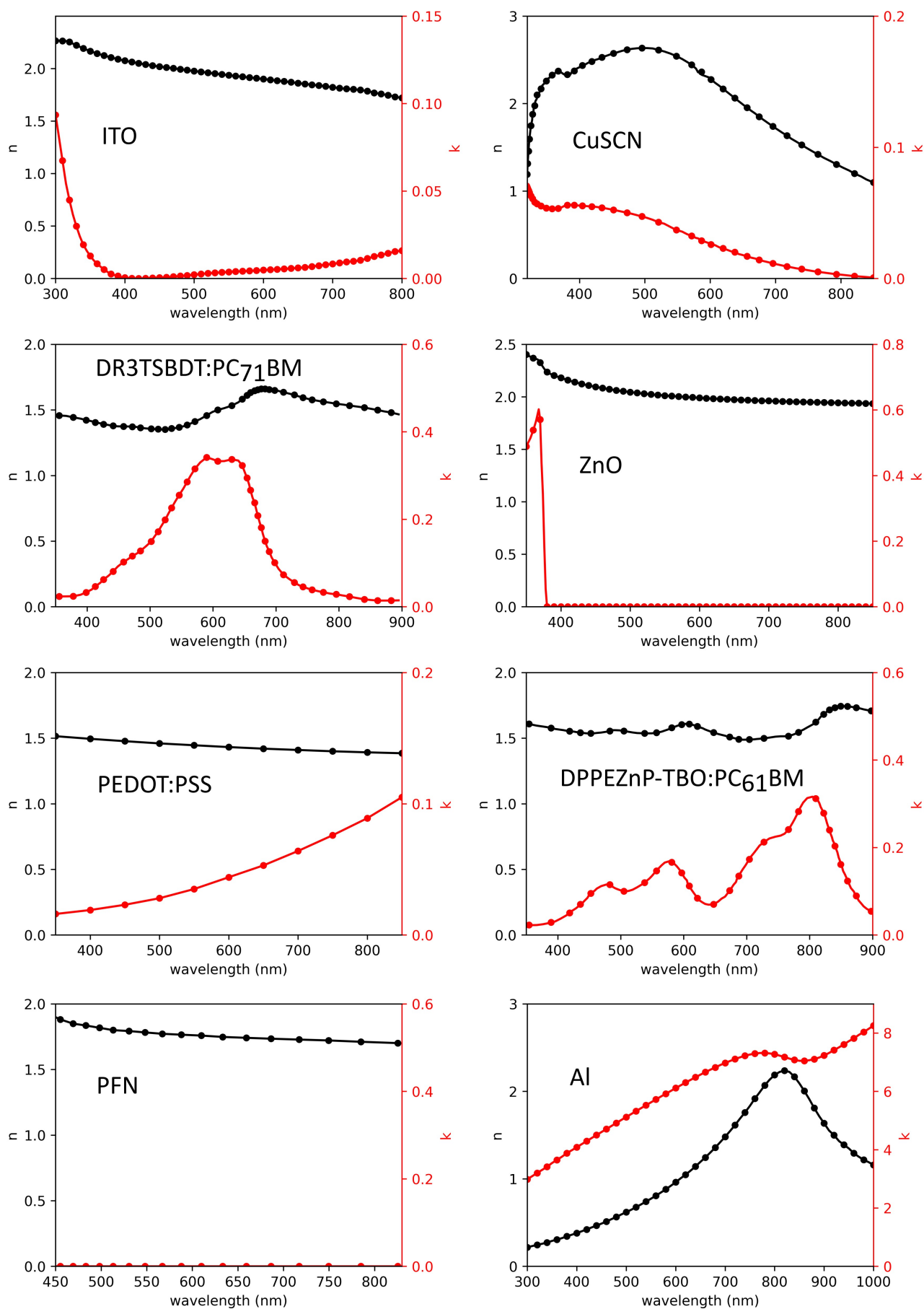
The refractive index  $n$  and extinction coefficient  $k$  of the materials included in the optical simulations were taken from the following references:

- ITO: `Setfos` material database, data originally from Woollam VASE.
- CuSCN: data from Ezealigo et al. [2020].
- DR3TSBDT:PC<sub>71</sub>BM: data from Kumari et al. [2017].
- ZnO: `Setfos` material database, originally from Sun and Kwok [1999].
- PEDOT:PSS: `Setfos` material database, originally from H.C. Starck, [www.hcstarck.com](http://www.hcstarck.com).
- DPPEZnP-TBO:PC<sub>61</sub>BM: data from Xiao et al. [2017].
- PFN: data from He et al. [2012] (supplemental information).
- Al: `Setfos` material database, data originally from Woollam.

In the cases where only absorption data was available, the wavelength-dependent refractive index was obtained from a Kramers-Kronig transformation. The resulting data is shown in Fig. S1. For the use with `Setfos`, the  $nk$ -data was interpolated/extrapolated to the wavelength grid of the solar illumination spectrum considered in the simulation, which is the ASTM G173-03 reference spectrum from <http://rredc.nrel.gov/solar/spectra/am1.5/> shown in Fig. S2.

## **2 ELECTRICAL MATERIAL PARAMETERS**

The labelling of the electrical material parameters is introduced in the main text. For the charge carrier mobilities, the same sources as for the optical data were used (see above). Intrinsic HOMO-LUMO levels were taken from the main reference Li et al. [2017]. For ZnO, parameters were taken from Fan and Lu [2005], except for the mobilities, for which lower values corresponding to the nanoparticle morphology of the interlayer are chosen. Regarding the electrode work functions, the literature value of 4.7 eV was used for ITO/PEDOT:PSS in the single junction configuration, while larger value of 4.97 eV in tandem due to CUSCN band tails Kim et al. [2016]; similarly, a value of 3.9 eV modified due to PFN for ideal band alignment was chosen for the Al work function. The values for defect density and capture rates were obtained from a fit to the experimental JV characteristics (marked by an asterisk in Tabs. S1 and S2). Additionally, a series resistance of  $1.2 \Omega \text{ cm}^2$  is assumed for the top cell in single junction configuration, while a shunt resistance of  $1.6 \text{ k}\Omega \text{ cm}^2$  is used for all cells. For the effective attempt frequency of the charge hopping at the recombination junction, a value of  $\mathcal{V} = 5 \times 10^{-5} \text{ m}^2/\text{s}$  is chosen.

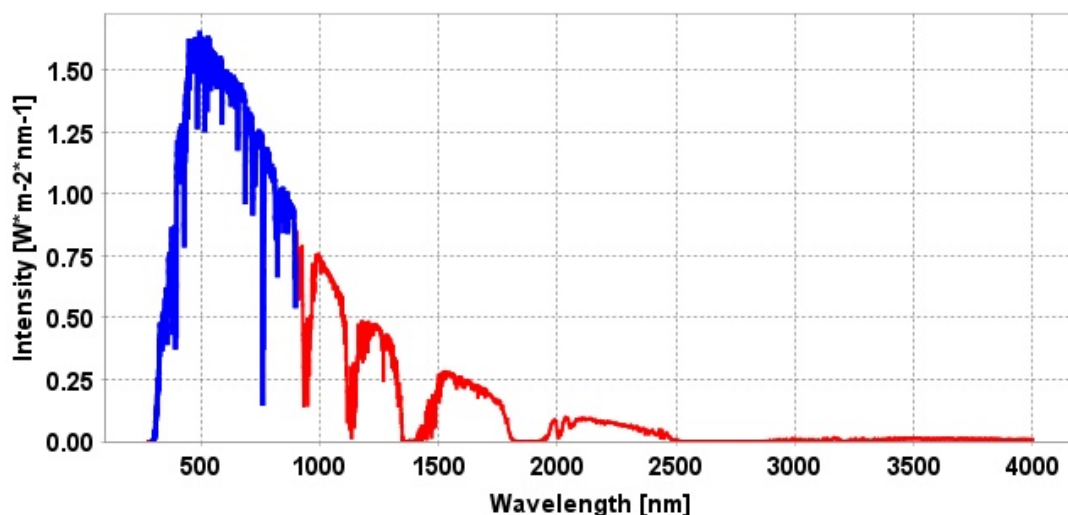
**Figure S1.** Refractive index and extinction coefficient data used in the optical simulation with Setfos.

**Table S1.** Electrical material parameters used for the simulation of the single junction cells.

|  | ITO | PEDOT:PSS          | DR3TSBDT:PC <sub>71</sub> BM | DPPEZnP-TBO:PC <sub>61</sub> BM | PFN              | Al  |
|--|-----|--------------------|------------------------------|---------------------------------|------------------|-----|
| WF $\Phi$ [eV]                               | 4.7 |                    |                              |                                 |                  | 3.9 |
| HOMO [eV]                                    |     | 4.9                | 5.07                         | 5.23                            | 7                |     |
| LUMO [eV]                                    |     | 3.6                | 4                            | 3.9                             | 3.9              |     |
| $\epsilon_r$                                 |     | 3.5                | 3.0                          | 3.5                             | 3.5              |     |
| $N_{0n}$ [cm <sup>-3</sup> ]                 |     | 10 <sup>21</sup>   | 10 <sup>21</sup>             | 10 <sup>21</sup>                | 10 <sup>21</sup> |     |
| $N_{0p}$ [cm <sup>-3</sup> ]                 |     | 10 <sup>21</sup>   | 10 <sup>21</sup>             | 10 <sup>21</sup>                | 10 <sup>21</sup> |     |
| $N_t$ [cm <sup>-3</sup> ]                    |     |                    | 10 <sup>12*</sup>            | 2.5×10 <sup>15*</sup>           |                  |     |
| $E_t$ [eV]                                   |     |                    | 0.5                          | 0.65                            |                  |     |
| $C_n$ [cm <sup>-3</sup> s <sup>-1</sup> ]    |     |                    | 10 <sup>-10*</sup>           | 3.3×10 <sup>-10*</sup>          |                  |     |
| $C_{np}$ [cm <sup>-3</sup> s <sup>-1</sup> ] |     |                    | 10 <sup>-10*</sup>           | 3.8×10 <sup>-10*</sup>          |                  |     |
| $\mu_n$ [cm <sup>2</sup> /Vs]                |     | 10 <sup>-4</sup>   | 2×10 <sup>-5</sup>           |                                 | 10 <sup>-5</sup> |     |
| $\mu_{0n}$ [cm <sup>2</sup> /Vs]             |     |                    |                              | 7.9×10 <sup>-4</sup>            |                  |     |
| $\mathcal{E}_{0n}$ [kV/cm]                   |     |                    |                              | 100                             |                  |     |
| $\mu_p$ [cm <sup>2</sup> /Vs]                |     | 2×10 <sup>-5</sup> | 1.6×10 <sup>-5</sup>         |                                 | 10 <sup>-7</sup> |     |
| $\mu_{0p}$ [cm <sup>2</sup> /Vs]             |     |                    |                              | 0.008                           |                  |     |
| $\mathcal{E}_{0p}$ [kV/cm]                   |     |                    |                              | 100                             |                  |     |
| $\eta_{\text{gen}}$                          |     |                    | 0.9                          | 1.02                            |                  |     |
| $N_{\text{acc}}$ [cm <sup>-3</sup> ]         |     | 10 <sup>19</sup>   |                              |                                 |                  |     |

**Table S2.** Electrical material parameters used in the simulation of the tandem cell (only parameters differing from subcell simulations shown).

|                                      | ITO  | CuScN                | DR3TSBDT:PC <sub>71</sub> BM | ZnO                | DPPEZnP-TBO:PC <sub>61</sub> BM |
|--------------------------------------|------|----------------------|------------------------------|--------------------|---------------------------------|
| WF $\Phi$ [eV]                       | 4.97 |                      |                              |                    |                                 |
| HOMO [eV]                            |      | 5.5                  |                              | 7.7                |                                 |
| LUMO [eV]                            |      | 1.8                  |                              | 4.1                |                                 |
| $\epsilon_r$                         |      | 5.1                  |                              | 8.5                |                                 |
| $N_{0n}$ [cm <sup>-3</sup> ]         |      | 10 <sup>21</sup>     |                              | 7.3 <sup>20</sup>  |                                 |
| $N_{0p}$ [cm <sup>-3</sup> ]         |      | 10 <sup>21</sup>     |                              | 2.8 <sup>21</sup>  |                                 |
| $N_t$ [cm <sup>-3</sup> ]            |      |                      | 3.3×10 <sup>13*</sup>        |                    | 2.4×10 <sup>14*</sup>           |
| $\mu_n$ [cm <sup>2</sup> /Vs]        |      | 10 <sup>-4</sup>     |                              | 1                  |                                 |
| $\mu_p$ [cm <sup>2</sup> /Vs]        |      | 0.1                  |                              | 0.1                |                                 |
| $\eta_{\text{gen}}$                  |      |                      | 1.3                          |                    | 1.28                            |
| $N_{\text{don}}$ [cm <sup>-3</sup> ] |      |                      |                              | 5×10 <sup>18</sup> |                                 |
| $N_{\text{acc}}$ [cm <sup>-3</sup> ] |      | 7.2×10 <sup>17</sup> |                              |                    |                                 |



**Figure S2.** AM1.5G solar spectrum (blue part considered in the simulation).

## REFERENCES

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