

Current efficiency of solar cells

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Structure (a-c) and AFM images (d-f) of Zn_2ZnTCPP , $\text{Cu}_2(\text{CuTCPP})$ and $\text{Cu}_2(\text{ZnTPyP})$ MONs used in this work, (g-i) scatter plots of the height of the MONs (in nm) vs the largest lateral dimension in nm.

(a) Current-voltage curves and (b) PCE averaged over 30 devices for each P3HT-MON-PC71BM combination are plotted as a function of the active layer composition, please consider the color code presented in the legend below the panels.

(a) External quantum efficiency (EQE) of the P3HT-additive-PC71BM devices without and with different ternary additives, and (b) thin-film absorption spectra of the pure MONs cast from ethanol.

These findings confirm that the MONs are photoactive in the blends, with the porphyrin units contributing to net light absorption. However, this benefit does apparently not result in a higher EQE. In contrast, the photoresponse in the primary absorption region of P3HT-PC71BM at 500 nm is weakened by 5% with the incorporation of $\text{Cu}_2(\text{ZnTPyP})$ and by 30% in the case of $\text{Cu}_2(\text{CuTCPP})$. This indicates charge carrier extraction issues, presumably due to changes in the active layer morphology and charge transport pathways within these devices - an observation that will be explored below.

(a) UPS (He I) valence band spectra of PEDOT:PSS without (bottom spectrum) and with different spin-coated MON additives. (b) Magnified region of the same spectra on logarithmic scale for inspection of the leading edge to derive the valence band maxima (VBM), indicated by the red arrows. For the spectrum of PEDOT:PSS the background (red dashed line) is indicated, revealing that there is occupied density of states straight up to the Fermi level (EF) at 0 eV.

The performance metrics of P3HT-PC71BM reference device and P3HT- $\text{Zn}_2(\text{ZnTCPP})$ -PC71BM device stored outside the glovebox at room temperature for 1 month were tested again (ESI Sect. 12, Fig. S42, Tables S8 and S9). The PCE of the devices after 1 month MONs were found to be 1.44% with MONs and 0.76% without the MONs. This indicates that the presence of the nanosheets has no detrimental effect on the stability of the devices over time and continue to offer improved power conversion efficiencies as compared to the devices without nanosheets.

Here we have shown that the choice of organic ligand and metal ion can have a significant difference in device performance, either enhancing ($\text{Zn}_2(\text{ZnTCPP})$), reducing ($\text{Cu}_2(\text{CuTCPP})$) or having no-effect ($\text{Cu}_2(\text{ZnTPyP})$)

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compared to reference devices without nanosheets. We then explored three different possible underlying explanations for the differences observed, examining how structural differences affect optical absorption, energy level alignment, and morphology.

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