

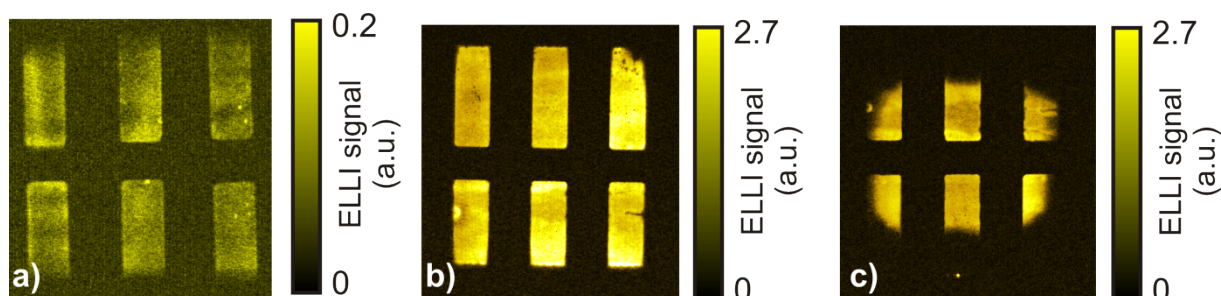
Electronic Supplementary Information (ESI)

## Air-processed organic tandem solar cells: Toward competitive operating lifetimes

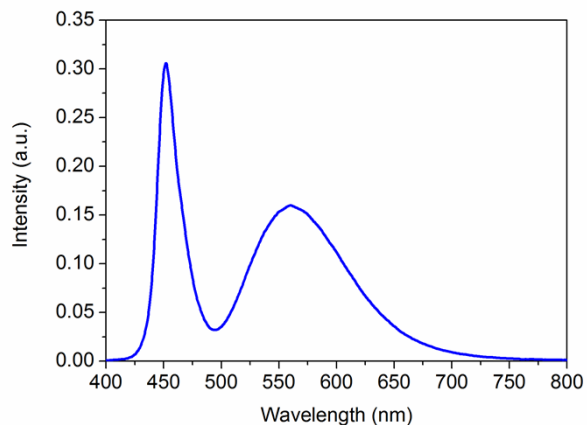
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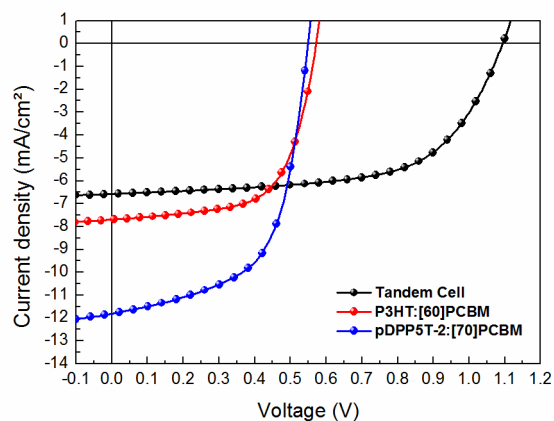
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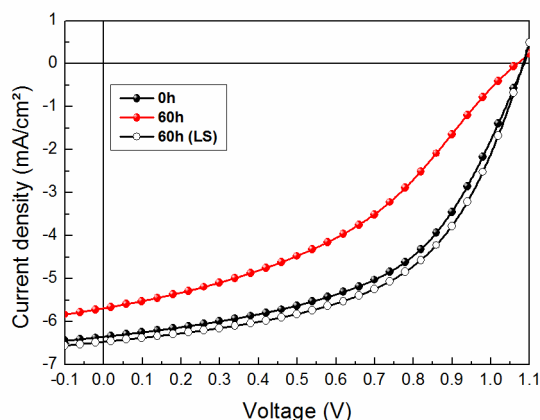
**Figure S1.** Electroluminescence lock-in (ELLI) imaging of photoaged OPV tandem cells. a) ELLI image of photoaged tandem cells after 2000 h of continuous white light illumination. No significant change in EL radiation was observed during the 2000 h of continuous irradiation, suggesting stable packaging. (b) and (c) show ELLI images of fresh and aged P3HT:PCBM single cells, respectively (see Figure 1 for device geometry). The aging process was accelerated by storing the sample in a climate chamber (dark) with 65°C/85% relative humidity for 168 h. After a storage time of 168 h, a clear disruption of the ELLI signal becomes apparent due to encapsulation failure (c). For ELLI imaging an Equus 327k NM infrared (IR) camera (IRCAM GmbH, Erlangen, Germany) was used, equipped with a cooled indium-gallium-arsenide (InGaAs) FPA detector (640x512). The spectral response of the detector is between 0.8  $\mu\text{m}$  and 1.8  $\mu\text{m}$ . The camera was run at a frame rate of 60 Hz and standard lock-in detection was applied. The cells were biased with 1.5 V in forward direction. For a detailed description we refer the reader to Hoyer et al.<sup>1,2</sup>



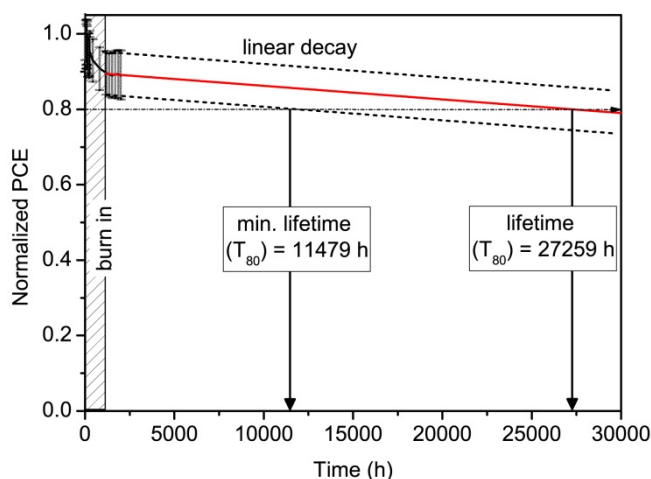
**Figure S2.** Spectral emission of the white light LEDs (BRIDGELUX - BXRA-30E0800-B-00) used for photodegradation of the tandem and single cells.



**Figure S3.** J-V characteristics of a representative tandem cell and their respective sub-cells (see Figure 1 for device geometry) before photoaging. The initial device performance of the tandem and single cells under AM1.5 conditions was  $4.4\% \pm 0.2\%$  (tandem),  $2.8\% \pm 0.1\%$  (P3HT), and  $4.1\% \pm 0.2\%$  (pDPP5T-2).

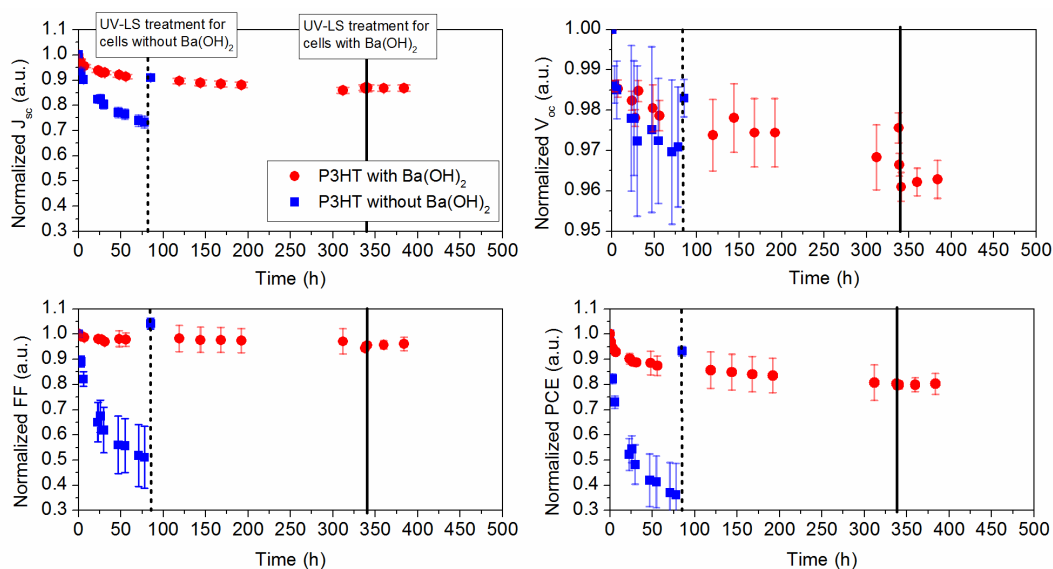


**Figure S4.** S-shape formation in the J-V performance of OPV tandem cells based on the active materials P3HT:PC[60]BM and pDPP5T-2:PC[70]BM. The device performance was probed immediately upon UV light soaking for 10 s (0 h, black curve), after 60 h under continuous photoaging (red curve) and after repeated UV treatment (60 h, black dashed curve; LS means after light soaking).



**Figure S5.** Extrapolated lifetime of inverted OPV tandem cells. Long-term PCE decay of inverted OPV tandem solar cells based on a P3HT:PC[60]BM front cell and a pDPP5T-2:PC[70]BM rear cell. Each data point represents an average value of 5 tandem devices. For estimating the accelerated lifetime, we applied a linear fit of the form  $y = 0.899x - 3.6 \times 10^{-6}$  to the data points following the burn-in period and extended the fit to where the efficiency drops

to 80% of the initial value (red line). For a minimum expectable lifetime of our cells we extrapolated the minimum (maximum) values of the error bars (dashed lines). The lifetimes were calculated considering an average 1500 hours of sunshine per year (central Europe). See main text for more details.



**Figure S6.** Photovoltaic device parameters of inverted P3HT:PCBM single solar cells with and without a  $\approx 20$  nm thick  $\text{Ba}(\text{OH})_2$  interfacial layer under continuous photoaging.

The device geometry was the same as in Figure 1 of the main text:

ITO/AZO/ $\text{Ba}(\text{OH})_2$ /P3HT:PCBM/PEDOT:PSS/Ag. At time  $t=0$  the devices were UV light soaked ( $\lambda_c = 365$  nm) for 10 s. Continuous photoaging occurred under white light illumination using high power LEDs without UV component (400 – 750 nm) at  $\approx 1000$   $\text{W}/\text{m}^2$ . UV light treatment was repeated after 78 h for cells without  $\text{Ba}(\text{OH})_2$  and after 340 h for cells containing a thin  $\text{Ba}(\text{OH})_2$  layer. Importantly, devices with  $\text{Ba}(\text{OH})_2$  feature significantly improved overall device stability. Moreover, no light soaking effect is apparent upon repeating the UV treatment as opposed to devices without  $\text{Ba}(\text{OH})_2$ . Each data point represents an average value of 5 solar cells and is normalized to  $t = 0$ . The photovoltaic parameters were extracted from J-V curves measured under an AM1.5 solar simulator at  $1000$   $\text{W}/\text{m}^2$ .

## References

- 1) U. Hoyer, L. Pinna, Th. Swonke, R. Auer, C. J. Brabec, T. Stubhan, and N. Li, *Adv. Energy Mater.* 2011, **1**, 1097–1100.
- 2) U. Hoyer, M. Wagner, Th. Swonke, J. Bachmann, R. Auer, A. Osvet, and C. J. Brabec, *Appl. Phys. Lett.*, 2010, **97**, 233303.