## **Supporting Information**

## Electrolytes as Cathode Interlayers in Inverted Organic Solar Cells: Influence of the Cations on Bias-Dependent Performance

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**Figure S1.** J-V curves for devices with EDTA-4H interlayer under (a) 1 V, (b) 2 V, and (c) 4 V bias for different time.



**Figure S2.** *J*-*V* curves for devices with EDTA-2H2Na interlayer under (a) 1 V, (b) 2 V, and c) 4 V bias for different time.



**Figure S3.** *J*-*V* curves for devices with EDTA-1H3Na interlayer under (a) 1 V, (b) 2 V, and c) 4 V bias for different time.



**Figure S4.** J-V curves for devices with EDTA-4Na interlayer under (a) 1 V, (b) 2 V, and (c) 4 V bias for different time.



**Figure S5.** Atomic force microscope (AFM) images of bare ITO surface and EDTA-X (X = nH(4-n)Na, n = 0, 1, 2, and 4) interlayers coated on ITO: (a) Bare ITO, RMS = 2.43 nm, (b) EDTA-4H, RMS = 2.52 nm, (c) EDTA-2H2Na, RMS = 2.49 nm, (d) EDTA-1H3Na, RMS = 2.79 nm, and (e) EDTA-4Na, RMS = 2.52 nm. (Size: 2.0  $\mu$ m × 2.0  $\mu$ m)



**Figure S6.** Ultraviolet photoelectron spectroscopy (UPS) of EDTA-nH(4-n)Na (3 mg/mL) coated ITO and bare ITO.



**Figure S7.** Variation of PCE with bias time for the devices with EDTA-nH(4-n)Na interlayers under (a) 1 V and (b) 4 V bias.





**Figure S8.** Current density of ITO/interlayer/Al devices with EDTA-nH(4-n)Na (a) n = 4, (b) n = 2, (c) n = 1, and (d) n = 0 under 2 V bias for 90 s.



**Figure S9.** J-V curves for devices with and without adding PCBA into active layers.

Figure S10. Decay of normalized PCEs of the non-encapsulated i-OSCs with EDTA-nH(4-n)Na (a) n = 4, (b) n = 2, (c) n = 1 interlayers in glovebox, and (d) the comparison of their decay rate.

