Supporting Information

**Interface engineering yields efficient perovskite light-emitting diodes**

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**1. Materials**

At 0°C, under continuous stirring for a duration of 2 hours, 4-fluorophenylmethylamine and methylamine were combined with equimolar quantities of aqueous HI (stabilized, sourced from Sigma-Aldrich) to synthesize 4-fluorophenylmethylammonium iodide (FPMAI) and methylammonium iodide (MAI). The FPMAI and MAI have recrystallized twice in N2 glovebox. Their ethanol solutions were slowly cool down at a rate of 5 °C/h for recrystallization purposes. After production of the large white crystals, they were washed with diethyl ether and subsequently dried at 60°C in a nitrogen glovebox. The lead iodide (PbI2) was purchased from Alfa Aesar. The PAI (99%), BAI (99%), and DAI (99%) all purchased from Xi'an Polymer Light Technology Corp.

**2. Perovskite precursor solution**

The 0.6 M MAPbI3 precursor solution was prepared using N,N-dimethylformamide (DMF) from Sigma-Aldrich through the equimolar blending of MAI and PbI2, to which FPMAI was then added at 20% molar excess relative to the MAPbI3 solution. The concentrations of PAI, BAI, and DAI were fixed at 10 mg/ml in isopropanol (IPA) purchased from (ACS, ≥99.5%). As a prerequisite for use, the precursor solutions underwent stirring for 3 hours at room temperature to validate appropriate blending.

**3. Device fabrication**

The initial step for the indium tin oxide (ITO) patterned glasses was washing with detergent, deionized water, acetone, and ethanol. The second step was drying at 70 °C. Afterward, ITO glass slides were exposed to UV light for 10 minutes before film deposition. Following that, the ITO substrates were coated with a thin layer of poly(N,N'-bis-4-butylphenyl-N,N'-bisphenyl)benzidine (poly-TPD). Here the final concentration was fixed at 6 mg mL-1 using a spin-coating technique. The spin coating speed to deposit the poly-TPD layer was fixed at 1000 rpm for 60 s and then annealed at all substrates at 150 °C for 20 minutes. Poly-TPD layers were treated with O2 plasma for 6 s to improve wetting. For the MAPbI3 perovskite layer deposition, the spin-coating rate was set at 5000 rpm for a total duration of 45 s. Upon the spin process stabilizing at 5000 rpm, a 70 μL toluene solution was dropped onto the spinning substrate after 4 seconds elapsed. All spin-coated MAPbI3 layers were later subjected to a 10-minutes post-annealing treatment at 70°C. Subsequently, these MAPbI3 perovskite emissive layers were post-treated with PAI, BAI, and DAI (10 mg/ml) dissolved in IPA separately and annealed again at the same temperature and time. The devices were finalized by depositing 2,2ʹ,2″-(1,3,5-benzinetriyl)-tris(1-phenyl-1-H-benzimidazole (TPBi), LiF: lithium fluoride (LiF), and aluminum (Al) layers of 40 nm, 1.2 nm, and 100 nm thicknesses through thermal evaporation.

**4. Characterizations of perovskite films**

The (Rigaku TTR-III) diffractometer was used in Bragg-Brentano parallel beam configuration with the standard Cu X-ray source to obtain X-ray diffraction (XRD) data for structural characterization. The scanning electron microscopy (SEM) analysis of without (control) and post-treated perovskite films was performed by field-emission SEM (GeminiSEM 500). Utilizing a 430 nm excitation wavelength, the photoluminescence (PL) and PLQY spectra were obtained by means of the Horiba Fluorolog-3 spectrometer joined with a Petite Integrating Sphere. The Horiba time-correlated single photon counting setup was used to perform time-resolved photoluminescence (TRPL) characterizations. A pulsed 369 nm laser diode excitation source from (NanoLED-Horiba), with an excitation intensity of approximately ~6 mWcm-2, was utilized to photoexcite the samples. Triple exponential fitting was utilized to fit the decays curves: decay(t) = , , where is the normalized coefficient and is the time constant, respectively.

**5. PeLEDs characterization**

PeLED measurements were performed in a nitrogen-filled glovebox using a custom motorized goniometer system including Keithley 2400 source meter (Keithley Instruments), FDS-100-CAL silicon photodiode (Thorlabs), Agilent 4140B picoammeter (Agilent Technologies), and UVN-SR spectrophotometer (StellarNet Inc.).



Figs. S1. (a-d) SEM images of perovskite films before and after post-treatment.

Table S1. Transient and steady-state optical properties of control and post-treated perovskite films with PAI, BAI, and DAI, respectively.

