Supporting Information

All-Solution-Processed Perovskite Light-Emitting Diodes with All

Metal Oxide Transport Layers

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Experimental Section

Device fabrication:

The PeLED devices are fabricated on the glass substrates with indium-doped tin oxide (ITO, 10 $\Omega \cdot \text{cm}^{-2}$) coated. The ITO substrates were sonicated sequentially in decon soap, distilled water, acetone, and isopropyl alcohol for 15 min each. The cleaned ITO substrates were then transferred into the UV-ozone chamber and treated for 15 min.

The preparation of nickel precursor solution, nickel formate dihydrate was dissolved in ethylene glycol, equivalents of ethylenediamine (Aldrich) was then added into the precursor prepared by fully stirring and last the completely dissolved solution with clarified purple color was filtered with 0.45 μ m nylon filters. The preparation of NiO_x electrode interlayer, the prepared solution was spin-coated on glass/ITO substrate at 4500 rpm for 90 s. Then, the substrate was annealed in atmospheric environment at 400 °C for 10 min to form the NiO_x interfacial layer on glass/ITO substrate.

The CsPbBr₃/BABr thin films were spin-coated from solutions with PEO and varied CsPbBr₃/BABr(1:0.3, 1:0.5, 1:0.8) concentrations in dimethyl sulfoxide (DMSO) solvent. The precursor solution was prepared by dissolving CsBr/PbBr₂/BABr and PEO in DMSO, respectively. Before spin coating, CsPbBr₃/BABr precursor solutions were prepared by mixing CsBr and PbBr₂ and BABr at different molar ratios in anhydrous DMSO to give a concentration of 200 mg mL⁻¹. PEO was dissolved in DMSO to give a concentration of 10 mg mL⁻¹. The perovskite solution and PEO solution were stirred at 60 °C overnight, and premixed at 2:1 volume ratio. Perovskite thin films were prepared by spin coating, the films were annealed at 80 °C for 10 min to evaporate the solvent. The effect of varied CsPbBr₃/BABr ratios on the crystal structure and optoelectronic properties are studied.

The ZnMgO nanoparticle was dissolved in ethanol and spin-coated onto the CsPbBr₃ serving as ETL. Al (100 nm) were thermally deposited sequentially under high

pressure. The active area of device is around 3.6 mm² defined by the overlapping area of ITO anode and Al cathode.

Device characterization:

The current–luminance–voltage measurement of the devices was carried on a computer-controlled Keithley 2400 source measure unit and Keithley 2000 Multimeter with a calibrated Si photodiode. The above measurement was carried out in a nitrogen-filled glovebox without encapsulation. EL spectra were collected by a spectrometer (Ocean Optics 2000) with an optical fiber in atmospheric environment without encapsulation and the EL emission spectrum assuming Lambertian profile.

Steady-state photoluminescence (PL):

PL spectra was recorded by a fluorescence spectrophotometer (Hitachi, F-4600).

X-ray diffraction (XRD):

The crystal structures of perovskite thin films were characterized by an X-ray diffractometer (Bruker, D8 Focus) using Cu K α (λ =0.15406 nm) as the X-ray source. Diffraction patterns were measured from 5-60° 2 θ at a scanning rate of 5° min⁻¹.



Fig. S1 2D and 3D AFM images of the perovskite films with different molar ratio of BABr:CsPbBr₃ in the precursor solution, respectively. The molar ratios are marked on the left images and roughness of the films are marked on the left sides of the 3D images, respectively. The topographic color-map is changed according to the color scale bars. All white scale bars are 2 μ m.



Fig. S2 SEM images of the perovskite films with different molar ratio of BABr:CsPbBr₃ in the precursor solution. The molar ratios are marked on the images. All white scale bars are 1 μ m.



Fig. S3 AFM images of the perovskite films without and with PEO in the precursor solution, respectively. The molar ratios and roughness of the films are marked on the images, respectively. All white scale bars are $2 \mu m$.



Fig. S4 Normalized PL spectra of perovskite thin films with different BABr:CsPbBr₃ molar ratios prepared on glass substrate and ITO substrate.



Fig. S5 Histograms of (a) peak current efficiency (CE) and (b) peak luminance of PeLEDs measured from 45 devices (BABr:CsPbBr₃ = 0.8:1). The average peak CE is 2.97 cd A⁻¹ with a low relative standard deviation (RSD) of 9.4%, and the average peak luminance is 14521 cd m⁻² with a low RSD of 11.3%.



Fig. S6 The EL spectra of PeLED devices based on BABr:CsPbBr₃ = 0.8:1 driven under different applied bias.



Fig. S7 Commission Internationale de l'Eclairage (CIE) color-space coordinates of the PeLED device based on BABr:CsPbBr₃ = 0.8:1.