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High performance organo-lead halide perovskite light-emitting diodes via surface passivation of phenethylamine



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ABSTRACT

One of the organometal halide perovskite materials, *i.e.* methylammonium lead bromide (MAPbBr₃), possesses interesting and incredible physicochemical properties, and shows great promising in light-emitting device. However, the poor morphology and high defect density properties of MAPbBr₃ films limit the performance of perovskite light-emitting diodes (PeLEDs). Here, high efficiency PeLEDs based on MAPbBr₃ is demonstrated by the incorporation of phenethylamine (PEA). We find that the introduction of PEA into the precursor of MAPbBr₃ reduces the grain size and increases the coverage of MAPbBr₃ films. In addition, it also passivates the surface defects of the grains, which all lead to high photoluminescence efficiency. In order to promote the charge injection into the MAPbBr₃ rapins, suppress the exciton quenching at the interface of perovskite film and hole transporting layer (HTL)/electron transporting layer (ETL), the device structure of "insulator-perovskite-insulator" is employed to further improve the performance. Our work presents an facile and robust route for high performance PeLEDs.

1. Introduction

Recently, organic–inorganic lead halide perovskite materials, with a three-dimensional ABX₃ structure (where A for methylamine $(CH_3NH_3^+, MA)$ or formamidine $(HC(NH_2)_2^+, FA)$ cation; B for Pb²⁺, Sn²⁺ or Ge²⁺ metal ion; X for I⁻, Br⁻, Cl⁻ halide ion [1,2]), have attracted considerable interests in optoelectronic applications owing to their interesting and incredible physicochemical properties [3–5], such as facile solution process ability [6–9], balanced charge transport properties with long diffusion lengths [10,11], tunable band-gaps [12–14], high carrier mobilities [15–17], and low non-radiative recombination ascribed to their unique defect chemistry [18,19]. Moreover, the demonstration of high photoluminescence (PL) quantum efficiency in the hybrid perovskite suggests effective light emitters for light-emitting diodes, and the optical absorbance can be easily tuned across the entire visible spectrum *via* halogen substitutions [20–23].

Since the first realization of perovskite light-emitting diodes (PeLEDs) at room temperature pioneered by R. Friend and his coworkers in 2014 [24], organic-inorganic lead halide perovskite materials have been at the basis of several breakthroughs in the development of light-emitting devices. So far, H. Cho et al. overcame the electroluminescence efficiency limitation of PeLEDs and made a breakthrough in current efficiency (CE) of the device up to $42.9 \text{ cd } \text{A}^{-1}$ and external quantum efficiency (EQE) to 8.53% within only one year [25]. However, the efficiency of PeLEDs is intrinsically limited by poor coverage and big grains of the perovskite film directly prepared by a one-step spin-coating method from its pure precursor solution, resulting in serious non-radiative leakage current losses in the PeLEDs. So far, a number of approaches have been developed to overcome those problems. In the preparation process of perovskite thin films, anti-solvents such as chlorobenzene, toluene, and diethyl ether are used to control the formation process of the perovskite film to achieve a uniform and

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smooth surface [26,27]. These interfacial treatments, however, are complex and delicate, which limits the reproducibility of the fabrication process of the perovskite film. In addition, two-step methods have been widely adopted to attain uniform perovskite films [28]. However, these methods could lead to reduced grain size and poor crystallinity of perovskite films, resulting in high defect density and thereby inferior luminescent properties. Therefore, preparation of smooth perovskite films with full coverage and passivation of the grain surface are still open problems to be solved for the high performance PeLEDs.

In this work, we introduce phenethylamine (PEA) into methylammonium lead bromide (MAPbBr₃) system to improve the performance of PeLEDs devices based on MAPbBr₃ thin film. PEA is found to effectively improve the coverage and passivate the surface defect of MAPbBr₃ film. By utilizing the device structure of "insulator-perovskiteinsulator" that we developed recently, an efficient green emission with luminance of 29268 cd m⁻² and the maximum current efficiency of 9.81 cd A⁻¹ with promising device stability can be achieved.

2. Experimental section

2.1. Perovskite precursor

A 38 wt% MAPbBr₃ perovskite precursor solutions (PbBr₂:MABr molar ratio = 1:1.5) were prepared by dissolving PbBr₂ (Aldrich, 99.999%) and MABr (Borun New Material Technology LTD) in anhydrous dimethylformamide (DMF), and then stirred at 60 °C for 2 h in a nitrogen-filled glove box. After that, 1 μ l, 3 μ l, 5 μ l, 10 μ l and 20 μ l PEA solutions (Acros Organics, 99%) were separately added into 1 ml of the above-mentioned MAPbBr₃ precursor solutions in which the final volume ratios of PEA solution to MAPbBr₃ precursor solution is 0.1%, 0.3%, 0.5%, 1% and 2%, respectively. After stirring at 60 °C for 1 h in a nitrogen-filled glove box, the hybrid precursor solutions were obtained.

2.2. Fabrication of PeLEDs

Firstly, indium tin oxide (ITO) patterned glass substrates with sheet resistance of about 15Ω /square were cleaned with deionized water and organic solvents, and then exposed to UV-ozone ambience for 5 min. Secondly, the substrates were transported into a vacuum chamber, and then 3 nm LiF (Sigma-Aldrich, 99.999%) layer was deposited onto the ITO-coated substrates via thermal evaporation under high vacuum $(< 3 \times 10^{-3} \text{ Pa})$ with the rate of about 0.2 Å/s monitored in situ by a quartz-crystal sensor. After deposition, the ITO/LiF substrates were transferred into a nitrogen-filled glove box in which the above-mentioned perovskite precursors were spin-coated (3000 rpm for 30 s) onto the substrates and then annealed at 80 °C for 15 min to form the perovskite layer. Finally, these substrates were transported into the vacuum chamber again, and then, LiF (8 nm)/Bphen (60 nm)/LiF (1 nm)/ Al (100 nm) were sequentially deposited onto the substrates under a pressure of 3×10^{-3} Pa at a rate of about 0.2 Å s⁻¹, 1 Å s⁻¹, 0.2 Å s⁻¹ and 5 Å s⁻¹, respectively. In our experiments, the light emitting devices have an effective area of about 12 mm².

2.3. Sample characterization

Film thickness of the samples was measured with ellipsometry (SE MF-1000, Korea). The surface of perovskite layer was investigated by scanning electron microscopy (SEM, Quanta 250, FEI). The crystalline structure of the perovskite layer on ITO substrate was determined by X-ray diffraction (XRD, D/MAX-2400, Rigaku, Japan) with Cu-K α radiation. The absorption and PL spectra were obtained by a UV–vis spectrophotometer (HITACHI U-3010, Japan) and a fluorescence spectrometer (Fluoromax-4 spectrofluometer), respectively. Time-resolved PL spectra were recorded with 50 ps time resolution using time-correlated single photon counting (TCSPC) system (FLS920 spectrometer) (excited by picosecond pulsed LEDs, pulse duration, < 850 ps, repetition rate,

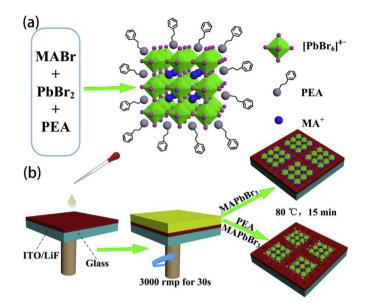


Fig. 1. (A) The design of MAPbBr₃ with chemically addressable ligands. The inorganic layers consist of corner-sharing lead bromide octahedra in which the black and pink balls represent Pb and Br atoms, respectively. The organic layers consist of MA⁺ cations and ligand PEA of blue and gray, respectively. (b) Schematic diagram of the MAPbBr₃ film formation process. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

10 MHz). The luminance-current-voltage (L-I-V) characteristics of the devices were measured using a computer-controlled source meter (Keithley 2602) and a calibrated silicon photodiode. The EL spectra were collected by a PR650 spectrometer. All light emitting measurements were carried out in a nitrogen-filled glove box at room temperature.

3. Results and discussion

By using one-step solution process, the schematic diagram of the MAPbBr₃ structural change after PEA introduction is shown in Fig. 1a. In the (PEA) (MA)PbBr₃ perovskite, small fraction of three-dimensional (3D) MAPbBr₃ cluster is protected by long-chain PEA ions, which greatly improves the stability of the MAPbBr₃ crystal structure. The discrete quantum level generated by this "quantum well" structure is reported to be more conducive to the selective carrier transport [5,29]. As shown in Fig. 1b, we fabricated the device by introducing different amounts of PEA into MAPbBr₃ precursor solution, and then spin-coating onto an indium tin oxide (ITO) substrate covered with a thin LiF layer followed by post-annealing (see Experimental Section).

The surface morphology and microstructure of the perovskite film have a critical impact on the performance parameters of the device. SEM images of MAPbBr3 films fabricated with different amounts of PEA in MAPbBr₃ precursor solution is shown in Fig. 2a-f. As shown in Fig. 2a, when no PEA is introduced, the perovskite film with large grains and low surface coverage can be observed. When the PEA ratio increases to 0.1%, the surface coverage of MAPbBr₃ film begins to increase, though the grain size is not found to reduce, as shown in Fig. 2b. Furthermore, when the PEA ratio increases to 0.3%, the surface coverage of resultant film is significantly improved and the crystal size is relatively reduced as shown in Fig. 2c. While the PEA ratio exceeds 0.3%, the surface coverage of MAPbBr₃ film gradually increases as shown in Fig. 2d-f. As depicted, the surface coverage of the MAPbBr₃ film is significantly increased with increasing dosage of PEA, especially, the surface coverage reaches 100% when the dosage of PEA is more than 1%.

The crystallinity of the perovskite films is also verified by XRD. As

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