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دیود نورگسیل پروسکایتی بر پایه $FAPbBr_3$

پریناز معززی[۱]، وحید احمدی[۱]، فرزانه عربپور رق‌آبادی [۱و۲]، بهرام عبدالهی نژند[۱]، مسعود پاینده

[۱]ایران، تهران، دانشگاه تربیت مدرس، دانشکده مهندسی برق و کامپیوتر، گروه پژوهشی اپتوالکترونیک و نانوفوتونیک

[۲]ایران، تهران، دانشگاه تربیت مدرس، دانشکده مهندسی شیمی، گروه فرایند

نویسنده مسئول: v_ahmadi@modares.ac.ir

چکیده- پروسکایت‌های هیبریدی آلی-معدنی در مطالعات سال‌های اخیر برای کاربردهای در افزاره‌های اپتوالکترونیکی توجه بسیاری را به خود جلب کرده‌اند. این مواد نیمه‌هادی دارای خواص نوری مناسبی از جمله طول نفوذ بالای حامل‌ها، بازترکیب نوری حامل‌ها، انتقال حامل خوب و خلوص نور بالا هستند که این خواص، برای ساخت افزاره‌های نورگسیل بسیار مطلوب می‌باشد. در این تحقیق، از پروسکایت فرمامیدینیوم برمایـد به‌علت بازترکیب نوری بالا، جهت ساخت دیود نورگسیل پروسکایتی استفاده می شود که این دیود نورگسیل با نور سبز که پیک آن در ۵۳۷ نانومتر است، در ولتاژ ۲/۷ ولت گسیل نور آن شروع می شود.

کلید واژه- دیودهای نورگسیل پروسکایتی، بازترکیب نوری، افزاره‌های اپتوالکترونیک

Perovskite light emitting diode based on $FAPbBr_3$

Parinaz Moazzezi[1] , Vahid Ahmadi[1], Farzaneh Arabpour Roghabadi[1,2], Bahram Abdolahi Nejand[2], Masoud Payandeh

[1] Optoelectronic and Nanophotonic Research Group, Faculty of Electrical and Computer Engineering, Tarbiat Modares University, Tehran-Iran.
[2] Faculty of Chemical Engineering, Tarbiat Modares University, Tehran, Iran

*Corresponding author: v_ahmadi@modares.ac.ir

Abstract- Recently, organometal halide perovskite has been attracted a lot of attention to be employed in optoelectronic devices. Perovskite desired properties like long carrier diffusion length, high radiative recombination, and pure luminescence introduced these materials as suitable candidates to use in optoelectronic devices. In this study, perovskite LED based on formamidinium lead bromide ($FAPbBr_3$) as the emitter layer is used. Perovskite LED emits green light with λ=537nm that starts emitting at 2.7V.

Keywords: Organic-inorganic halide perovskite, Perovskite light-emitting diode, Optoelectronic device

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1. Introduction

Organic-inorganic halide materials are emerging as highly remarkable semiconductors during the last few years. Their superior properties such as their long carrier diffusion length, high photoluminescence quantum yield, and tuneable bandgaps over the entire visible spectral extent enable us to use them in electronic and optoelectronic devices. Furthermore, halide perovskite can be processed from precursor solutions at low temperature to form crystalline direct-bandgap semiconductors with shallow traps. The electroluminescence (EL) of these devices is limited by non-radiative recombination at interfaces and grain boundaries, which is associated with defects and high leakage current due to non-uniform perovskite layer [1,2].

In the early perovskite LEDs, $CH_3NH_3PbI_3$ and $CH_3NH_3PbBr_3$ were used as emitter layer for red and green emission, respectively, with narrow FWHM~20 nm, while quantum dot LEDs yield an FWHM around 30 nm. Thus, perovskite LEDs had greatly attracted a lot of attentions. After reporting these organic-inorganic based devices, inorganic perovskites such as $CsPbX_3$ (X=Cl, Br, I) were also used as emitter layers in LEDs to provide the fabrication of devices with higher thermal stability and higher photoluminescence quantum efficiency [3].

As already reported, $HC(NH_2)_2PbBr_3$ ($FAPbBr_3$) shows superior carrier lifetime and diffusion length compared to $MAPbBr_3$ in the form of single crystal and solution processed films, that reveals its greater potential to be employed in optoelectronic devices. Moreover, FA^+ has a larger ion radius than MA^+ in the A-site of perovskite as demonstrated in Fig (1). It leads to a higher tolerance factor in the range between 0.9 and 1 that is preferred at room temperature for desired crystal stability. Therefore, $FAPbBr_3$ is a suitable choice as a green light emitter in perovskite LEDs [4].

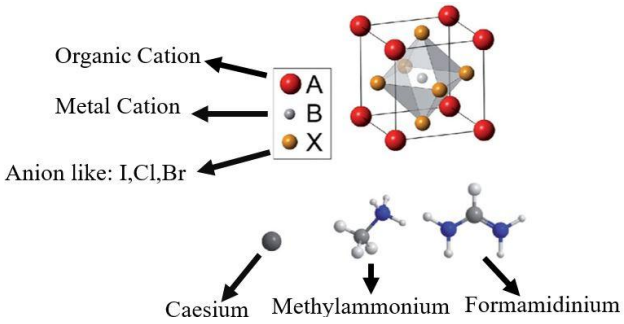


Figure 1: The crystalline structure of perovskite material.

In this study, perovskite LEDs with ITO/PEDOT: PSS/FA based perovskite/ZnO/Ag structure is fabricated as shown in Fig. (2), using one-step solution processed deposition method. To improve the PL property of the perovskite layer, also its morphology is engineered.

2. Method

2.1. Synthesis of $CH(NH_2)_2Br$ (FABr)

To synthesize FABr, formamidinium acetate powder is dissolved in hydro bromic acid (HBr) (48 wt. %). After addition of acid, the solution is stirred for 10 minutes at 50℃. Upon drying at 100℃, a yellow-white powder is formed that is washed with diethyl ether and recrystallized twice with ethanol. Before using, the powder is dried for 24 h in a vacuum oven [5].

2.3. Fabrication method

- The glass/ITO substrate with 15 Ω resistivity is sequentially washed with deionized water and ultrasonic bath, then oxygen plasma treated.
- PEDOT: PSS as hole transporting layer is spin-coated at 4000 rpm and anneal at 140 ^{0}C for 10 min.
- Perovskite precursor containing FABr and $PbBr_2$ dissolved in DMF is deposited using one–step method at 4000 rpm and annealed at 70℃.
- In the next step, the electron transporting material, ZnO nanoparticles, is spin coated at 6000 rpm.
- Finally, Ag is deposited as the device contact.

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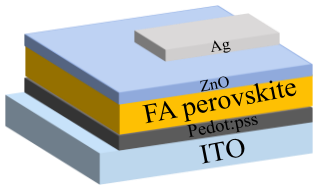


Figure 2: Schematic of the structure of the fabricated PLED.

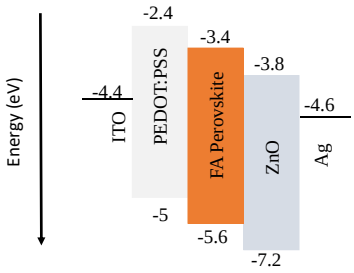
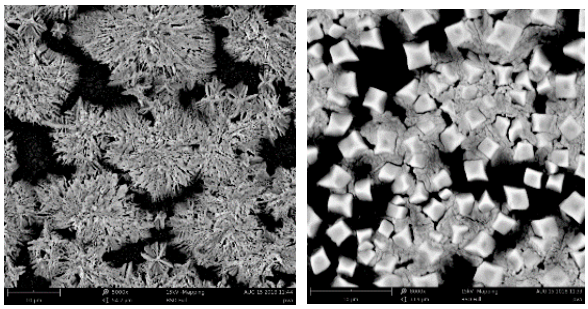


Figure 3: The energy level diagram of ITO/PEDOT: PSS/$FAPbBr_3$ perovskite/ZnO/Ag device.

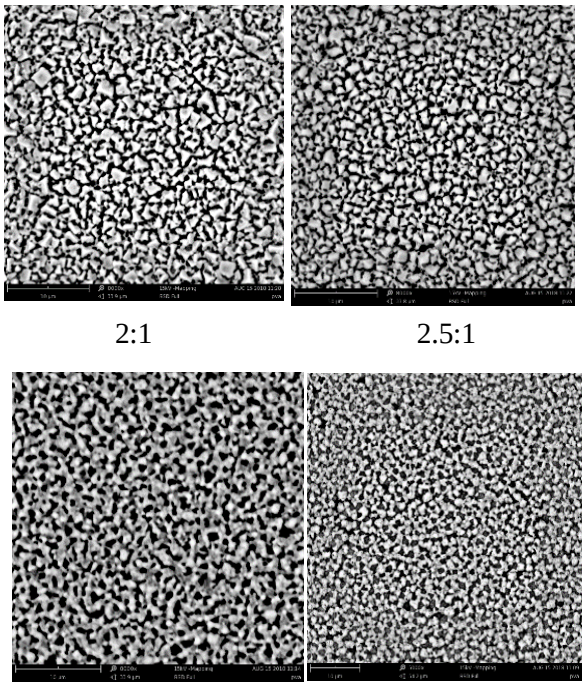
3. Result and discussion

The energy level diagram of ITO/PEDOT: PSS/$FAPbBr_3$ perovskite/ZnO/Ag device is shown in Fig. (3). as observed in Fig. (4), morphology of the perovskite layer is strongly dependent on the ratio of formamidinium bromide and $PbBr_2$. The desirable grain size for more efficient charge carrier confinement and radiation recombination in LEDs is achieved when, 3:1 ratio of FABr: $PbBr_2$ is used (Fig.4).

As SEM images show in Fig. (4), within increasing the FABr ratio precursor, grains become smaller and the surface coverage enhances. However, the presence of a high amount of excess FABr in the layer leads to the loss of the stability of the layer.



1:1 1.5:1



2:1 2.5:1

3:1 3.5:1

4:1 4.5:1

Figure 4: SEM images of the perovskite layers synthsized from precursors containing different ratios of FABr: $PbBr_2$, (SEM images scale bar is 10µm).

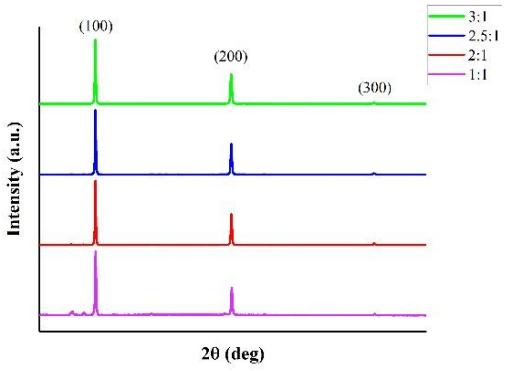


Figure 5: XRD patterns of $FAPbBr_3$ synthsized from precursors containing different ratios of FABr: $PbBr_2$

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The X-ray diffraction (XRD) patterns in Fig. (5) depict that the FA-based perovskite has a $Pm\bar{3}m$ cubic crystal structure that shows no dependency on the precursor ratio [6].

Fig. (6) presents the absorption and PL spectra of the perovskite film synthesized from the precursor with 3:1 ratio. The small grains formed in this film provides a desirable PL performance which its peak locates at 530 nm.

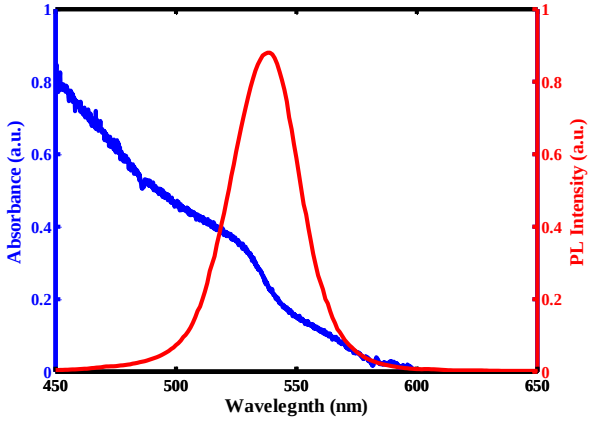


Figure 6: PL spectrum and absorption spectrum of $FAPbBr_3$

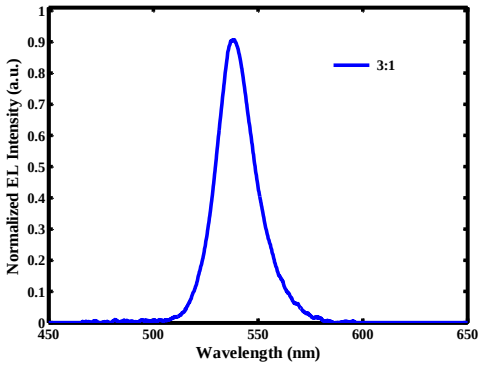


Figure 7: EL spectrum of the PeLED with 3:1 ratio of FA-perovskite precursor as emission layer

EL spectrum of the fabricated device is presented in Fig. (6). Remarkably, the device starts its emission at 2.7V at λ=537 nm with FWHM of 20 nm.

4. Conclusion

In summary, the effect of the various ratios of precursors on the morphology and crystalline structural of the $FAPbBr_3$ layer was investigated. It was depicted that the 3:1 ratio was the best ratio with suitable morphology. It provided the fabrication of high efficient perovskite LED that started its emission at 2.7 V (at λ=537 nm with FWHM of 20 nm).

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