

Improved performance of Si-based top-emitting organic light-emitting device using MoO_x buffer layer

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ARTICLE INFO

Article history:

Received 4 November 2009

Received in revised form 2 January 2010

Accepted 16 January 2010

PACS:

78.60.Fi

73.40.Lq ; 78.66.Qn

72.80.Cw ; 72.80.Le

Keywords:

OLED

Top-emitting

Si

Work function

Buffer layer

ABSTRACT

Highly efficient Si-based top-emitting organic light-emitting device (TOLED) using MoO_x buffer layer is demonstrated. With tris(8-hydroquinoline) aluminum as emitting and electron-transport layer, the p-Si/MoO_x based TOLED shows a maximum luminous efficiency of 1.1 cd/A and a power efficiency of 0.68 lm/W, which are almost double those (0.64 cd/A and 0.34 lm/W) of p-Si/SiO₂ based TOLED. Moreover, in comparison with the widely used thermally grown SiO₂ buffer layer, MoO_x can be deposited by conventional evaporation technology and thereby simplifying fabrication process.

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1. Introduction

Squeezing light out of Si has been attracted considerable attention for revolutionizing Si-based optoelectronic applications [1]. However, Si has indirect bandgap and thus quite inefficient band-to-band radiative electron–hole recombination. In recent years, the research interest of Si-based light emission is focused on the combination of Si and organic electroluminescence (EL) [1,2]. This hybrid technology gains many advantages such as no lattice mismatch between Si and organic materials, simple fabrication process, and good compatibility with modern Si technology [2,3]. In the early pioneering works [2–4], the efficiency of Si-based organic light-emitting device (OLED) is extremely low and the reasons for the poor performance remain unclear. Since Qin et al. [5] reported highly efficient Si-based top-emitting OLED (TOLED) using an ultra-thin buffer layer of thermally grown SiO₂, intensive investigation of

Si-based TOLED has been carried out and received increasing attention thereof [6–9]. Nowadays, researchers become more optimistic about Si-based TOLED, because this will open up an alternative route to the integration of optical devices with Si chips, including active matrix OLED (AM-OLED) displays.

Generally, two methods are widely used for realizing Si-based TOLED: (i) Si is directly used as anode, and (ii) Si coated with metal is used as electrode. For the latter, the metal determines carrier-injection characteristics and thereby device performance, while the Si only served as a substrate. For the former, the Si properties play a key role in determining the electro-optical characteristics. This structure allows the elimination of at least one mask for fabricating an AM-OLED, which simplifies the fabrication process and reduces manufacturing cost.

In Si-based TOLED, it is well established that the surface modification of Si is indispensable to improve device efficiency [4–5,8–10]. Buffer layers (e.g., FeCl₃, V₂O₅, MoO_x and WO₃) have been used in OLED with indium-tin-oxide (ITO) anode may also be employed for tuning carrier-injection characteristics of Si anode. MoO_x is superior to other choices of buffer layer in several respects: [11] (i) easy handling compared with the corrosive and optically absorbing FeCl₃; (ii) nontoxicity and high transmittance in comparison with V₂O₅; (iii) moderate evaporation temperature (much

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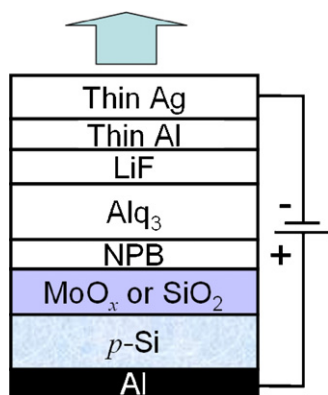


Fig. 1. Schematic structure of Si-based TOLEDs.

lower than WO_3) facilitates deposition process. On the other hand, most reported Si-based TOLEDs are realized using SiO_2 buffer layer [3,5–10]. SiO_2 must be obtained from other additional equipment, which will increase the cost and create contamination. In this article, we report the improved performance of Si-based TOLED using MoO_x buffer layer.

2. Experimental details

The p -Si wafer with (100) orientation and electrical resistivity of $10\ \Omega\text{cm}$ was served as substrate and anode. Al ohmic contact was formed in the backside. The TOLED was fabricated on the front side of Si under a vacuum of 3×10^{-4} Pa. MoO_x (2 nm) as a buffer layer and N,N' -bis(naphthalen-1-yl)- N,N' -bis(phenyl) benzidine (NPB, 60 nm) as a hole-transport layer were deposited subsequently, followed by 50 nm tris(8-hydroquinoline) aluminum (Alq_3) as an emitting and electron-transport layer. Trilayer LiF (0.3 nm)/Al (3 nm)/Ag (18 nm) semitransparent cathode [12] was deposited and patterned through a shadow mask to define the emitting area. Another Si-based TOLED using thermally grown SiO_2 (~ 2 nm) [5,6] as buffer layer was also fabricated for comparison. Fig. 1 shows the schematic structure of Si-based TOLEDs. We refer to the p -Si-based TOLED with MoO_x (or SiO_2) buffer layer as p -Si/ MoO_x (or p -Si/ SiO_2) device. The layer thickness of deposited materials was monitored *in situ* using an oscillating quartz thickness monitor. The deposition rate for organic materials was $\sim 1\ \text{\AA}/\text{s}$. The current–voltage–luminescence (I – V – L) characteristics were measured with a Keithley 2400 Source Meter and a Minolta LS-110 Luminescence Meter. The EL spectra were measured using a PR-650 Spectra Scan. Contact potential difference measurements were carried out as depicted in our early works [13].

3. Results and discussion

Fig. 2 shows the luminous efficiency–current density (η_l – J) and power efficiency–current density (η_p – J) characteristics of p -Si/ MoO_x and p -Si/ SiO_2 devices. It can be seen that the p -Si/ MoO_x device shows a maximum luminous efficiency of 1.1 cd/A and a power efficiency of 0.68 lm/W, which are almost double those (0.64 cd/A and 0.34 lm/W) of p -Si/ SiO_2 device. This indicates that MoO_x is more efficient than SiO_2 for enhancing device efficiency. We also noted that the luminous efficiency (0.64 cd/A) of p -Si/ SiO_2 device is slightly lower than that (~ 0.8 cd/A) of reported p -Si/ SiO_2 based TOLEDs with the same organic layers [6,9]. Such difference may be resulted from the following two factors: (i) the reported efficiency in literatures [6,9] is obtained using p -Si anode with an optimized resistivity ($40\ \Omega\text{cm}$), and (ii) the difference in semitransparent cathode, i.e., the transmittance of LiF (0.3 nm)/Al (3 nm)/Ag

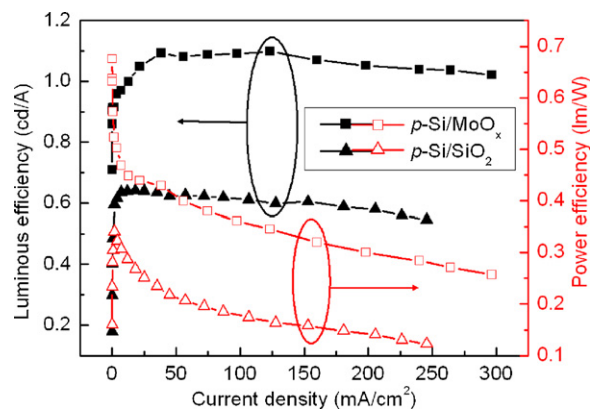


Fig. 2. η_l – J and η_p – J characteristics of p -Si/ MoO_x and p -Si/ SiO_2 devices.

(18 nm) semitransparent cathode is $\sim 30\%$ [12] while that of Yb (4 nm)/Au (15 nm) is $\sim 60\%$ [6]. However, the maximum luminous efficiency (1.1 cd/A) and power efficiency (0.68 lm/W) of p -Si/ MoO_x device are much higher than those (~ 0.8 cd/A and ~ 0.3 lm/W) of reported p -Si/ SiO_2 TOLEDs with the same organic stacks [5,6,9]. This indicates that MoO_x is an efficient buffer layer for tuning hole-injection characteristics of Si anode.

The J – V and L – V characteristics are depicted in Fig. 3 which clearly demonstrates that p -Si/ MoO_x and p -Si/ SiO_2 devices exhibit the same trend of J – V – L characteristics. This implies that the hole-injection characteristics from p -Si anode to NPB can be effectively controlled via the buffer layer of MoO_x just like via SiO_2 . But MoO_x is superior to SiO_2 in enhancing luminance and reducing driving voltage [Fig. 3].

It is well known that the work function (WF) of Si anode plays a key role in tuning carrier-injection characteristics and thus carrier balance within device [4,10]. To clarify the contribution of MoO_x and SiO_2 to the improvement of device performance, we measured the relative WFs of p -Si/ MoO_x and p -Si/ SiO_2 as a function of standing time after surface treatment [Fig. 4]. It is clear that MoO_x and SiO_2 modification can considerably enhance the WF of p -Si anode (enhanced by ~ 0.6 eV). The enhanced WF results in reduction of hole-injection barrier-height, which promotes carrier balance and improves device efficiency. On the other hand, heavily doped Si usually exhibits metallic properties. With the passivation of an ultrathin buffer layer, the modified-Si shows semiconductor properties which play a crucial role in tuning carrier-injection characteristics in Si-based TOLED [3,10]. In other words, the interfacial electronic structure of anode contact with the organic layer (NPB in this study) is an important factor controlling the hole injection [4]. SiO_2 is an insulating material, the semiconductor properties of

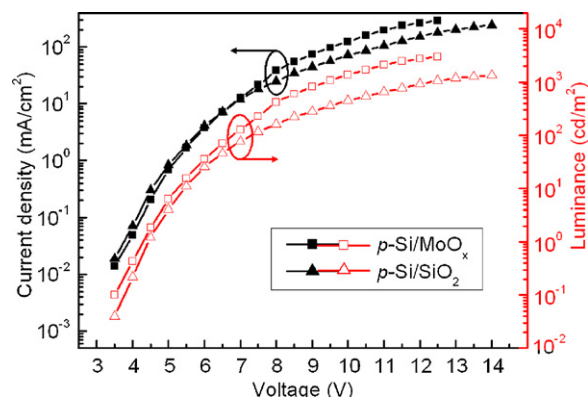


Fig. 3. J – V and L – V characteristics of p -Si/ MoO_x and p -Si/ SiO_2 devices.

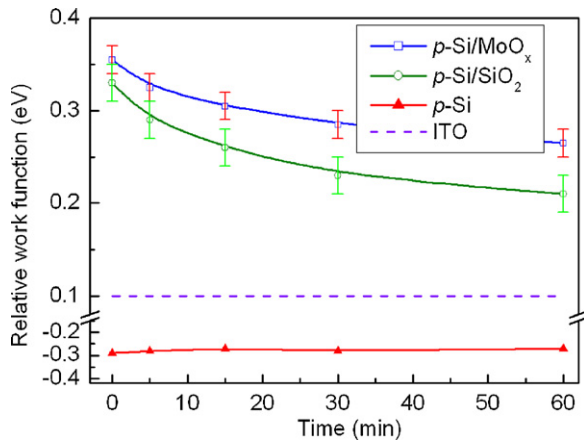


Fig. 4. The relative WFs of p -Si/ MoO_x and p -Si/ SiO_2 as a function of standing time after surface treatment. The characteristics of ITO and p -Si (without treatment) are also incorporated for comparison.

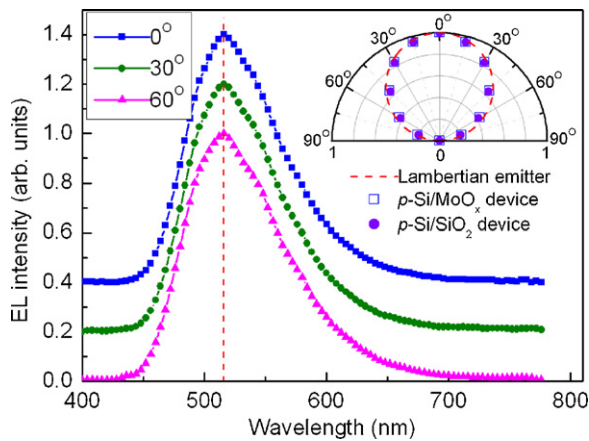


Fig. 5. EL spectra (normalized) of p -Si/ MoO_x device at viewing angles of 0° , 30° and 60° . For clarity, some curves have been shifted vertically. (Inset) The polar plots of emission intensities (normalized to the 0° intensities) of p -Si/ MoO_x device compared with p -Si/ SiO_2 device and Lambertian emitter.

SiO_2 -passivated Si are resulted from the trade-off between metallic Si and insulating SiO_2 [6,9]. While MoO_x is a semiconductor material with appropriate WF and superior hole-injection ability [11], the hole-injection characteristics can be predominantly controlled by MoO_x itself. The difference in surface electronic structure of p -Si/ MoO_x and p -Si/ SiO_2 may account for the difference of device performance. The p -Si/ MoO_x device shows higher current density in comparison with p -Si/ SiO_2 device at the same voltage ($>6\text{V}$) [Fig. 3], as both devices share the same cathode and organic stacks, the higher current density indicates an enhanced hole injection. Furthermore, the luminance of p -Si/ MoO_x device is much higher than that of p -Si/ SiO_2 device [Fig. 3], suggesting the enhanced hole injection contributes to better carrier balance and thereby promoting device efficiency. Therefore, MoO_x -modification is proven to be more favorable to improving EL performance. The WFs of p -Si/ MoO_x and p -Si/ SiO_2 decrease slightly with the increase of standing time after surface treatment [Fig. 4]. This is due to the surface electronic

structures of p -Si/ MoO_x and p -Si/ SiO_2 are influenced by the ambient conditions. Further studies are required to clarify the detailed mechanism. In addition, MoO_x is deposited under high vacuum condition which is free from contamination. This to great extent contributes to the improvement of device performance.

Fig. 5 shows the measured EL spectra (normalized) of p -Si/ MoO_x device at viewing angles of 0° , 30° and 60° off the surface normal. It can be seen that the EL spectra do not show blueshift with the increase of viewing angles. In the forward direction, the EL spectrum peaks at 516 nm with a full-width at half-maximum of 88 nm, giving green emission with Commission Internationale d'Eclairage (CIE) color coordinates of (0.278, 0.549). With increasing viewing angles, the CIE color coordinates show negligible variation. For example, at the viewing angle of 60° off the surface normal, the CIE color coordinates only shift to (0.280, 0.529). In addition, the polar plots of emission intensities (normalized to the 0° intensities) of p -Si/ MoO_x and p -Si/ SiO_2 devices are almost Lambertian distribution [inset in Fig. 5]. All these phenomena indicate that p -Si/ MoO_x device exhibiting weak microcavity effect as a result of low reflectivity of Si.

4. Conclusions

MoO_x and SiO_2 modification can considerably enhance the WF of p -Si anode, which accounts for the excellent EL performance of Si-based TOLED. MoO_x is proven to be more efficient than SiO_2 in improving device performance, the efficiencies of p -Si/ MoO_x device are almost double those of p -Si/ SiO_2 device. Moreover, MoO_x can be deposited by conventional evaporation technology, which simplifies the fabrication process and thus reduced manufacturing cost.

Acknowledgements

This work was supported by the National Natural Science Foundation of China (60577041, 60777018 and 60776040), 863 project (2008AA03A336), Shanghai Science and Technology Committee (06DZ22013), Shanghai Leading Academic Disciplines (S30107), and State Key Laboratory for Mesoscopic Physics (School of Physics, Peking University).

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