Efficient White PIN OLED Structures with Internal and External Light Outcoupling Enhancement

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ABSTRACT

Bottom and top emission white and monochrome PIN OLED structures with efficient outcoupling method based on vacuum-processable materials are reported. In combination with external microlens array film, a power efficiency of 60 lm/W at 1000 cd/m² with colour coordinates of 0.47/0.43 and CRI of 87 has been demonstrated.

1. INTRODUCTION

Organic light emitting diodes (OLEDs) are nowadays common in small display applications, and are expected to enter the large display market in a short time. In the area of general lighting, there has been an increasing number of demonstrators based on bottom and top emission white OLEDs. However, in addition to high production cost, also the power efficiency and in some cases the colour appearance still need to be improved in order that OLEDs become competitive with the other established technologies. Much of the current research activities are focused on development of emitter systems and charge transport materials that should allow higher internal quantum efficiencies and lower driving voltages. On the other hand, improved extraction of light from OLEDs promises a huge potential for efficiency enhancement.

For white OLEDs it is estimated that less than 30% of the generated light is coupled out into air. Most of the generated light is trapped in wave guided organic modes since the refractive index of organic layers and ITO is higher than that of glass substrate. The light that is propagating at the interface of organic layers and cathode is partly lost by dissipations in the metal cathode due to surface-plasmons. The third part remains trapped inside the glass substrate due to the difference in refractive index between glass and air.

As seen from the discussion above, the desired outcoupling method should ideally affect both the organic, surface-plasmon and substrate modes in order to maximize the outcoupling efficiency. In the past, a number of different approaches have been tested [1]. They involve modification of substrate surface, e.g. by using microlens arrays (MLA), photonic crystals, high-n glass substrates [2,3], scattering layers between substrate and anode [4], etc. However, most of these approaches either do not offer high efficiency enhancement over the broad range of wavelengths, or technologically they are not easy to implement.

In this paper, we report on recent progresses in efficiency enhancement with vacuum-processable materials NET-61 and NLE-17. By incorporating a layer of NET-61 in the electron transport layer of OLED stack, a considerable improvement of efficiency of white and monochrome bottom emission stacks has been demonstrated. In top emission stacks, NLE-17 cap layer on top of the semi-transparent cathode not only enhances efficiency, but also results in very stable colour coordinates over all viewing angles.

An alternative approach investigated recently involves a nanoparticle scattering layer between glass substrate and ITO. However, the efficiency enhancement achieved was smaller than in the approach based on vacuum-processable materials. In addition, the method suffers from processing issues and has a small potential for industrial applications.

2. RESULTS

2.1 Bottom emission OLEDs with NET-61

Recently we have reported on high efficiency improvement in single-unit and two-unit stacked (tandem) bottom emission white OLEDs by using NET-61 [5]. The reference devices and the devices with NET-61 have been processed on the standard ITO substrate with an active area of about 2x2 cm². The structure of the devices is shown in Fig. 1.

The one-unit stack is based on SFC fluorescent blue and orange emitters. Compared to the optimized reference stack, device A containing NET-61 shows an enhancement of external quantum efficiency of 47%. The power efficiency of device A is 31 lm/W at a brightness of 1000 cd/m² with colour coordinates of 0.48/0.42. The efficiency could be further improved by attaching an external MLA on the surface of the substrate for extraction of substrate modes. In this way the power efficiency of device A was increased to 37 lm/W at 1000 cd/m² (0.48/0.42).

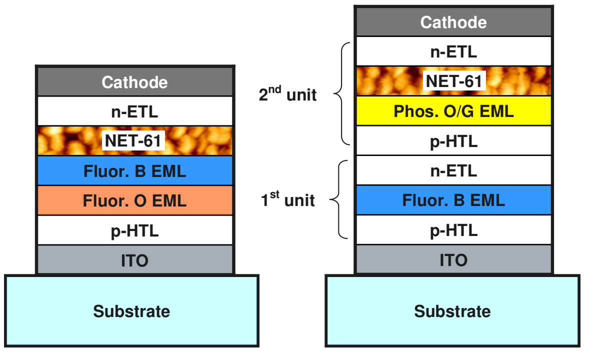

Fig. 1 Structure of single-unit white stack of device A (left) and tandem white stack of device B (right).

By optimizing the emitter systems we have been able to shift the colour coordinates to 0.43/0.40, which is within the DOE Energy Star tolerance quadrangle for 3000 K. Due to the change in the spectrum, the power efficiency achieved with an external MLA film was reduced to 34 lm/W at 1000 cd/m². Compared to the optimized reference stack, the total enhancement of quantum efficiency achieved by combination of NET-61 and an MLA film is 1.84. The stack shows the same extrapolated lifetime at 1000 cd/m² as the reference stack.

The highest efficiency reported so far had been achieved in a two-unit white stack consisting of a phosphorescent orange-green and a fluorescent blue unit. The power efficiency of the optimized reference sample is 27 lm/W at 1000 cd/m² (0.40/0.38). With NET-61 the efficiency was increased to 39 lm/W at 1000 cd/m² (0.43/0.40) for device B. By attaching an external MLA film, efficiency of device B was increased to 50 lm/W at 1000 cd/m² (0.46/0.41).

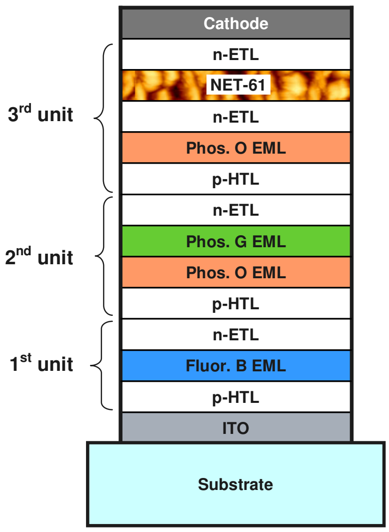

Fig. 2 Structure of 3-unit white stack of device C.

Recently we have been able to achieve even a higher efficiency by incorporating NET-61 into a 3-unit stacked white architecture. The stack consists of a fluorescent blue, phosphorescent orange-green and a phosphorescent orange unit (Fig. 2). A layer of NET-61 has been incorporated in the 3rd unit in between two n-doped electron transport layers (ETL). Compared to the optimized reference device, the power efficiency has been increased from 38 lm/W (0.41/0.42) to 51 lm/W at 1000 cd/m² (0.45/0.43) for device C containing NET-61. By using an external MLA film, the power efficiency has been further increased to 60 lm/W at 1000 cd/m² (0.47/0.43). The colour coordinates of device C with an MLA film are within the DOE Energy Star tolerance quadrangle for 2700K and colour rendering index (CRI) is 87. The corresponding emission spectrum measured in an integrating sphere is shown in Fig. 3. The stack shows an extrapolated T50 lifetime of 90,000 h at a luminance of 1000 cd/m² with an MLA film.

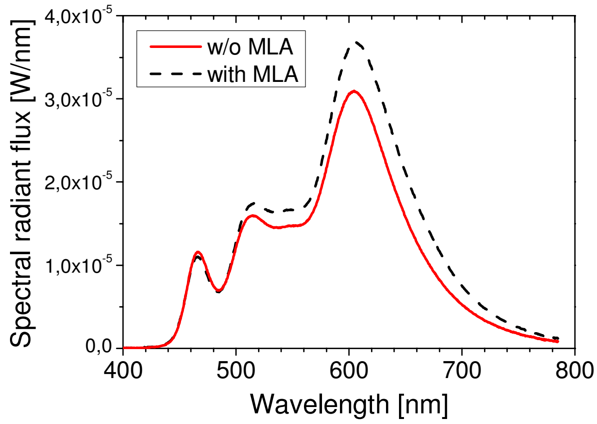

Fig. 3 Emission spectrum of device C at 1000 cd/m² without an external MLA film and with an MLA film attached.

In addition to white stacks, monochrome stacks have also shown large enhancement of efficiency with NET-61. Table 1 lists external quantum efficiencies achieved in case of a fluorescent green stack. With NET-61 external quantum efficiency is enhanced from 8.5% for reference device to 11.1% for device D containing NET-61, which presents an enhancement of 31%. By using an MLA film, efficiency of device D is further increased to 13.7%. A similar enhancement with NET-61 has been also achieved for monochrome blue and red stacks, which makes NET-61 suitable also for RGB displays.

In case of all above mentioned devices, a single layer of NET-61 has evaporated either between two n-doped ETLs or directly on hole blocking layer. The evaporated NET-61 forms crystallites on the underlying layer. The morphology and therefore the scattering properties can be controlled by changing evaporation parameters such as nominal layer thickness and evaporation rate. An SEM image of a device with NET-61 is shown in Fig. 4.

Due to NET-61, the interface between organic layers and cathode is not smooth, but shows a wavelike shape.

Table 1 External quantum efficiencies (EQE) in case of a fluorescent green stack. All values are at a constant current density of 3.5 mA/cm² corresponding to a luminance of about 1000 cd/m².

	Reference	Device D
EQE	8.5%	11.1%
EQE with MLA	12.6%	13.7%
Voltage	2.8 V	2.8 V

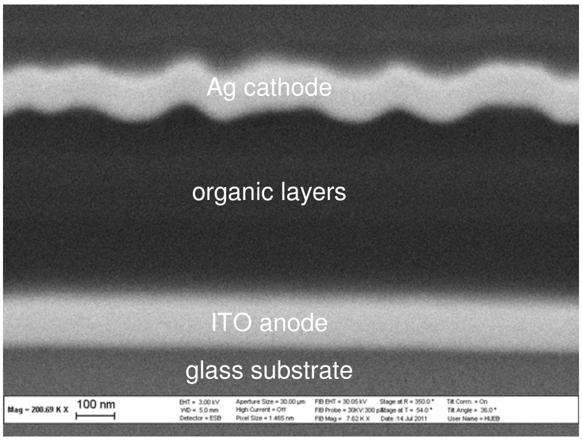

Fig. 4 SEM image of a cross-section of an OLED device with NET-61.

A silver cathode has been used in all stacks with NET-61. In comparison to other metals tested, highest efficiencies have been achieved with silver as cathode. In off-state, devices with NET-61 show frosted appearance in comparison to mirror-like appearance of reference devices. Due to the reduced reflectivity, the enhancement with an external MLA film is somewhat lower than for the reference devices. Despite the roughness, the I-V characteristic in the case of all stacks with NET-61 is not changed.

2.2 Mechanism of light extraction with NET-61

The frosted appearance of the cathode suggests that the improvement in efficiency with NET-61 is due to internal scattering at the rough organic-cathode interface. In order to distinguish between the contribution of organic and surface-plasmon modes, an experiment on a substrate with a high refractive index (n=1.8) has been performed. In this case the organic modes are almost completely extracted into the glass substrate, since the refractive index of high-n glass is comparable to that of organic layers and ITO. The contribution of the remaining surface-plasmon modes depends on the distance between the cathode and the recombination zone (d_C). Since the surface-plasmon losses decrease exponentially with d_C, the amount of light extracted into the substrate should increase for larger d_C.

Reference devices and devices E containing NET-61 have been processed with different ETL thicknesses. A single-unit fluorescent-phosphorescent white stack has been used on a high-n substrate with ITO. Based on Novaled PIN OLED® technology, ETL thickness variation provides an easy way to vary distance d_C without changing electrical properties of devices.

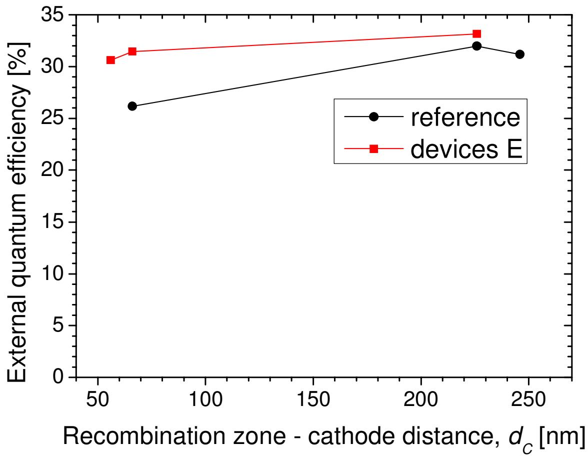

Fig. 5 Dependence of external quantum efficiency on recombination zone-cathode distance for reference devices and devices E with NET-61. The devices have been processed on a high-n substrate.

Fig. 5 shows external quantum efficiency as a function of d_C for the reference devices and devices E containing NET-61. In order to account for the substrate and air modes, external quantum efficiencies have been measured by attaching a large high-n half-sphere lens to the substrate. An optically matching high-n immersion oil has been used between the glass substrate and the lens.

As expected, with increasing d_C the efficiency of the reference devices increases due to reduced surface-plasmon losses [2]. On the other hand, only a very small increase has been observed in case of devices E. This suggests that reduction of surface plasmon losses in case of devices E accounts for the observed efficiency enhancement for small d_C.

In case of a standard-n (n=1.5) substrate, however, efficiency with NET-61 is considerably improved even for large d_C. Since for large d_C mainly organic modes are trapped, it can be concluded that NET-61 also extracts a part of organic wave guided modes. In conclusion, the experiments on high-n and standard-n substrate suggest that NET-61 helps to extract both surface-plasmon and organic modes trapped in the OLED stack.

2.3 Top emission OLEDs with NLE-17 and NET-61

Compared to bottom emission stacks, top emission OLEDs typically show stronger cavity effects. A thin semi-transparent metal layer is commonly used as a top contact, which together with a metal anode further increases the cavity effects. As a result, the cavity supports only a narrow range of wavelengths in forward direction. With increasing angle the cavity lengths becomes more suitable for longer wavelengths, therefore the devices exhibit a strong colour strongly shifts with the viewing angle.

A significant decrease in colour shift with viewing angle and increase in efficiency has been demonstrated by using NLE-17 scattering layer on top of the semi-transparent cathode [6]. By applying this approach to a tandem white stack, a power efficiency of 37 lm/W at 1000 cd/m² with colour coordinates of 0.45/0.41 and CRI of 75 has been achieved [5].

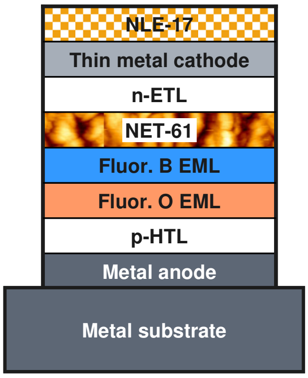

Fig. 6 Structure of top-emission single-unit two-colour fluorescent white stack with NET-61 and NLE-17 cap layer.

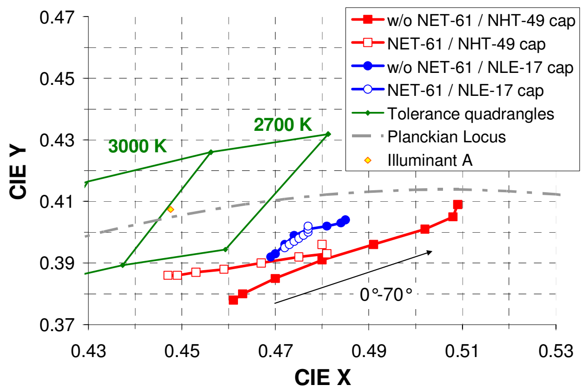

Fig. 7 Dependence of colour coordinates on viewing angle (0°-70°) for a single-unit white top-emission OLEDs with NHT-49 and NLE-17 cap layers, and with and without NET-61.

Recently we have demonstrated a further decrease of the colour shift by combining NLE-17 cap layer with NET-61 in the stack. The stacked used is based on fluorescent blue and orange emitters (Fig. 6). The colour shift with viewing angle is shown in Fig. 7. For devices without NET-61, the NLE-17 cap layer results in a much smaller colour shift compared to devices with a standard cap layer. In both cases, additional NET-61 layer reduces the colour shift.

3. SUMMARY

In the paper we have demonstrated high enhancement in efficiency by using vacuum-processable scattering material NET-61. By modifying the organic-cathode surface, light outcoupling in different white and monochrome bottom emission stacks could be improved. The method is fully compatible with standard vacuum fabrication technology and can be combined with external outcoupling solutions. By combining NET-61 with an MLA, external quantum efficiency could be enhanced by 84%. In a three-unit white stack, a power efficiency of 60 lm/W at 1000 cd/m² with colour coordinates of 0.47/0.43 has been achieved.

Power efficiency and colour shift with angle have been considerably improved in case of top emission OLEDs on metal with NLE-17 scattering cap layer. In a tandem white stack a power efficiency of 37 lm/W at a luminance of 1000 cd/m² with colour coordinates of 0.45/0.41 and CRI of 75 has been achieved.

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REFERENCES

[1] K. Saxena, V. K. Jain, and D. S. Mehta, Opt. Mater. 32, 221 (2009).

[2] S. Mladenovski, K. Neyts, D. Pavičić, A. Werner, and C. Rothe, Optics Express, Vol. 17, Issue 9, pp. 7562-7570 (2009).

[3] S. Reineke, F. Lindner, G. Schwartz, N. Seidler, K. Walzer, B. Lüssem, K. Leo, Nature, Vol 459, pp. 234 – 237 (2009).

[4] Y.-S. Tyan, Y.Q. Rao, X.F. Ren, R. Kesel, T. R. Cushman, W. J. Begley, N. Bhandari, SID 09 Digest, pp. 895-898 (2009).

[5] T.W. Canzler, S. Murano, D. Pavičić, O. Fadhel, C. Rothe, A. Haldi, M. Hofmann, Q. Huang, SID 11 Digest, pp. 975-978 (2011).

[6] J. Birnstock, T.W. Canzler, M. Hofmann, Q. Huang, T. Romainczyk, SID 10 Digest, pp. 774-777 (2010).