Characteristics performance and analysis of positional variation in charge generation layer to enhance performance of OLED.

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Research Article

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Abstract

In this paper, a highly efficient charge generation layer (CGL) based blue organic light emitting diode (OLED) is proposed. The proposed device contains a CGL which comprises of two materials 1,1-bis[(di-4-tolyamino)phenyl]cyclohexane (TAPC) and 1,4,5,8,9,11-hexaazatriphenylene-hexacarbonitrile (HAT-CN) which act as hole and electron injectors, correspondingly. The CGL in the proposed device is placed outside of the emissive layer which provides better luminescence and current as compared with four other CGL based devices D_2, D_3, D_4 and D_5 where CGL is utilized below cathode, above anode, near both electrodes (cathode and anode) and inside the emissive layer, respectively. The proposed device exhibits noteworthy results as peak values of current and luminescence are achieved as 0.44 A and 3636.3 cd/m^2, correspondingly. The obtained luminescence is about 16.8, 2.3, 1.7, 3, 1.6 times improved on comparing with D_1, D_2, D_3, D_4 and D_5. Furthermore, thickness optimization of the proposed device is also outlined. The optimized device showed the maximum luminescence of 4670 cd/m^2.

1. Introduction

Over the past few years, the researchers have shown their enormous interest in developing full colour high resolution displays which also attain better lifetime [1–3]. These organic material-based displays contain very attractive properties such as ultra-thin thickness, flexibility, easy fabrication, light weight, wide viewing angle, fast response etc. [4–7]. With these salient features, OLEDs have established their separate place in the display market which is gaining lot of attention of consumers nowadays. Despite having many attractive features, there is still need of improvement in the performance of these devices in terms of luminescence, current density, lifetime etc [8]. Although various researchers have incorporated many additional layers in OLEDs such as hole/electron injection layer, hole/electron transport layer, blocking layer etc. These layers have contributed a lot in order to enhance the performance parameters of organic LEDs [9, 10]. But, nowadays, some novel advanced layers which are mixed interlayer [11], charge generation layer [12], and spacer layers [13] have come into the picture to further refine OLED’s performance. These layers are being widely utilized by the researchers in recent time.

Maurya et al. [14] discussed an OLED device on which the positional analysis of mixed interlayer is performed. In this research, it is noticed that luminescence and current density are improved by changing the position of the layer towards the cathode. The peak luminescence and maximum current density are obtained as 17139 cd/m^2 and 84.6 mA/cm^2, correspondingly. Moreover, Liu et al. [15] inserted a mixed interlayer of poly(9-vinylcarbazole) (PVK) and poly[(9,9-dioctylfluorenyl-2,7-diyl)-co-(4,4′-(N-(4-s-butylphenyl)diphenylamine)) (TFB) between emissive layer and PEDOT:PSS layer. The proposed device exhibited maximum power efficiency of 47.2 lm/W and external quantum efficiency (EQE) of 18.86%. On the other hand, Ying et al. [16.] utilized a spacer layer with the materials 1-bis[4-(N,N-di(4-tolyl)amino)phenyl]-cyclohexane/2,6-bis[3-(9H-carbazol-9-yl)phenyl]pyridine/2,4,6-tris[3-(diphenylphosphinyl)phenyl]-1,3,5-triazine (TAPC-26DCzPPy-PO-T2T). The power efficiency and EQE are achieved as 76 lm/W and 24%, respectively.
In this paper, charge generation layer (CGL) is utilized for the improvement of luminescence and current density of OLED device. CGLs play a very important role to enhance overall device performance. In emissive layer after the recombination, only 20% light can be extracted out of the device, rest is lost due to surface Plasmon and optical losses. With the help of CGL, this lost part of light can be employed which automatically improves the device performance in terms of efficiency and EL emission [17]. These layers are the combination of two different layers (n type & p type) and act almost like electrodes. The charge carriers are produced depending upon the external applied voltage and injected to the emissive layer where the recombination takes place and light is produced [18].

Various researchers have studied CGL to refine the performance of organic LEDs [19–23]. In 2016, Kim et al. [24] discussed the emission properties of blue fluorescent multi-tandem organic LEDs using CGL of materials Al/MoOx. The maximum quantum efficiency and current efficiency are obtained as 25.6% and 46.6 cd/A, correspondingly. Addition to this, Yuan et al. [25] fabricated three OLEDs (red, green and blue) based on heterojunction CGL of HAT-CN/TAPC and improved the EL efficiencies of the fabricated devices which contain the balanced injection of electrons and holes. Moreover, Jetly et al. [17] utilized C70/Pentacene as CGL and designed a high-performance OLED. The reported CGL shows organic photovoltaic behaviour and the device parameters such as luminescence and current density are achieved as 15630 cd/cm² and 15.63 cd/A, respectively. Herein, a highly efficient organic LED is proposed using a charge generation layer which is a combination of two materials which are 1,1-bis[(di-4-tolyamino)phenyl]cyclohexane (TAPC) and 1,4,5,8,9,11-hexaazatriphenylene-hexacarbonitrile (HAT-CN). The CGL is placed outside the emissive in the proposed device. Incorporation of CGL in OLED provided better charge injection which results in higher current density and better luminous property. Additionally, this paper also compares five different CGL-based devices in terms of luminescence and current density.

This complete paper is distributed in total 5 sections. Section 1 provides the introduction which discusses various OLED devices and the important role of CGL in OLEDs. Section 2 is about the performance analysis of blue organic LED. Afterwards, Section 3 compares four various CGL based OLED devices and outlines their performance parameters curves. Moreover, Section 4 represents the proposed CGL based blue OLED. In this section, thickness reduction of charge generation layer is incorporated to achieve the optimized results of the proposed device. Eventually, Section 5 highlights the conclusion of the paper.

2. Characteristic Analysis and Parameters Extraction of Blue OLED

In order to design the proposed device using CGL, firstly a reference device [26] which is a multi-layered blue organic LED (D₁) without having CGL is validated. In this device, seven layers of different categories such as cathode, anode, electron transport layer (ETL), hole transport layer (HTL) and electron emissive layer (EML) are utilized as shown in Fig. 1. The length and width of the device structure are 400 and 1000 µm, respectively. The materials along with their thickness for these layers are illustrated in Table 1 below. The fabrication process starts with the deposition of an organic HTL of 90nm i.e. PEDOT:PSS (poly(3,4-ethylenedioxy thiophene):polystyrene sulfonate) using spin-coating technique on the top of anode layer. Afterwards, the thermal evaporation method is considered for the incorporation of the remaining layers.
above PEDOT:PSS. A hole transport layer of the material NPB (N,N'-Bis(naphthalene-1-yl)-N,N'-bis(phenyl) benzidine) having thickness of 60nm is placed on the top of PEDOT:PSS. Thereafter, an emissive layer of DPVBi (4,4'-Bis(2,2-diphenylvinyl)-1,1'-biphenyl) with 5% of BCzVBi (4,4'-Bis(9-ethyl-3-carbazovinylene)-1,1'-biphenyl) and with the thickness of 20nm is deposited. This layer is utilized for the generation of blue emission. Now, the deposition of 30nm BPhen (4,4'-Bis(9-ethyl-3-carbazovinylene)-1,1'-biphenyl) takes place which acts as combined layer of HBL (hole blocking layer) and ETL. Eventually, a cathode layer is placed which contains 1nm Lithium Fluoride and 150nm Aluminium.

<table>
<thead>
<tr>
<th>Layer</th>
<th>Material</th>
<th>Thickness (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Anode</td>
<td>ITO</td>
<td>20</td>
</tr>
<tr>
<td>HTL1</td>
<td>PEDOT:PSS</td>
<td>90</td>
</tr>
<tr>
<td>HTL2</td>
<td>NPB</td>
<td>60</td>
</tr>
<tr>
<td>EML</td>
<td>DPVBi &amp; BCzVBi</td>
<td>20</td>
</tr>
<tr>
<td>ETL</td>
<td>BPhen</td>
<td>30</td>
</tr>
<tr>
<td>Cathode</td>
<td>LiF</td>
<td>1</td>
</tr>
<tr>
<td>Cathode1</td>
<td>Al</td>
<td>150</td>
</tr>
</tbody>
</table>

In OLED, the generation of light is achieved with two important processes: 1) formation of charge carriers, and 2) recombination of charge carriers. For this, a model i.e. Langevin's recombination rate is applied in Silvaco Atlas 2-D device simulator for the validation of the reference device. The expression for the model is as follows:

$$R_L(n, p) = r_L(x, y, t) (np-n_i^2)$$

Here, $$r_L$$, $$n$$, $$p$$ and $$n_i$$ denote the recombination rate coefficient, electron concentration, hole concentration, and intrinsic concentration, respectively. Addition to this, to simulate the carrier’s transportation in a device structure Poole-Frenkel mobility model is utilized which was given by Frenkel in 1938. This model provides the results under specified boundary conditions. This model is given as:

$$\mu(E) = \mu_0e^\left[\frac{-\Delta}{kT}+(\frac{\beta}{kT}-\gamma)\sqrt{|E|}\right]$$

where $$\mu$$, $$\mu_0$$ and $$E$$ are field dependent mobility, zero field mobility, and electric field, correspondingly. Besides this, $$\Delta$$, $$k$$, $$T$$, $$\beta$$ and $$\gamma$$ represent activation energy at zero electric field, Boltzmann constant, effective temperature for charge carriers, hole Pool–Frenkel factor and fitting parameter, respectively. The device structure which is plotted using Silvaco is depicted in Fig. 2.
The curves above are showing the performance parameters such as current, current density and luminescence of blue OLED (D₁). The comparison of experiment and reference data of D₁ is drawn in Fig. 3 (a). From the figure, it is clear that there is less than 5% deviation in current parameter as reference and experiment devices contain 0.025 and 0.026 A, respectively. Also, Fig. 3 (b) is highlighting the combined curve for luminescence and current density. The maximum luminescence and current density are achieved as 215 cd/m² and 6.5 mA/cm², correspondingly.

3. Performance Analysis with CGL at Different Position for OLED

As discussed in Section 1, CGL is very useful for improving the concentration of charge carriers which can further enhance the luminous properties and current of OLED device. In this section, positional analysis using CGL is performed on D₁ device keeping same materials and thickness. Here, four blue OLED D₂, D₃, D₄ and D₅ are considered in which a 20nm CGL of TAPC (top layer) and HAT-CN (bottom layer) materials is incorporated on different positions as depicted in Fig. 4 (a), (b), (c) and (d). The combined layer of HAT-CN and TAPC act as electron and hole injectors, respectively.

The LUMO level of HAT-CN is approximately 5.5 ev, which is matching the HOMO level of TAPC i.e. also about 5.5 ev. This results in easy transfer of electrons from HOMO of TAPC to LUMO of HAT-CN which significantly increases the charge carriers in CGL [27]. In devices D₂ and D₃, CGL is utilized below the cathode and above the anode, correspondingly. On the other hand, in device D₄, it is placed near both electrodes like above the anode and below the cathode. Furthermore, in device D₅, CGL is situated inside the emissive layer in a manner as depicted in Fig. (d). The thickness of CGL in D₅ is taken as 40nm instead of 20nm due to their structural changes. All these four devices (D₂-D₅) are compared based on current density and luminescence with reference device D₁.

<table>
<thead>
<tr>
<th>Device</th>
<th>Current (A)</th>
<th>Luminescence (cd/m²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>D₁ (Reference)</td>
<td>0.026</td>
<td>215.9</td>
</tr>
<tr>
<td>D₂</td>
<td>0.193</td>
<td>1592.3</td>
</tr>
<tr>
<td>D₃</td>
<td>0.256</td>
<td>2112.2</td>
</tr>
<tr>
<td>D₄</td>
<td>0.145</td>
<td>1198.4</td>
</tr>
<tr>
<td>D₅</td>
<td>0.276</td>
<td>2277.2</td>
</tr>
</tbody>
</table>

Table 2
Comparison of Current and Luminescence of various blue OLEDs

Figure 5 (a) & (b) are highlighting current and luminescence parameters of all the devices (D₁-D₅). As shown in Fig. 5, device D₅ is exhibiting the best performance among other devices as in D₅ CGL is placed
inside of emissive layer which improves the recombination of charge carriers. The reason behind this is that in D₅ top layer (of TAPC) of CGL is acting as hole injector and bottom layer (of HAT-CN) is behaving like electron injector. Now, holes in top layer of CGL will be attracted towards cathode and will reach to emissive layer. Similarly, electrons in bottom layer of CGL will get attracted towards anode and reach to emissive layer as CGL in D₅ is surrounded by the emissive layer only. These charge carriers are mixed up with charge carriers which have travelled from cathode and anode side. This automatically enhances the recombination rate of the device. The maximum current and luminescence are obtained as 0.27 A and 2277.2 cd/m², correspondingly as depicted in Table 2.

4. Proposed Device

As shown in section 3, among all five devices (D₁ - D₅), D₅ is exhibiting remarkable performance. In this section, a high-performance blue OLED is proposed by reversing the position of CGL and emissive layer in device D₅ as illustrated in Fig. 6. Also, the additional modification is that in D₆ position of top and bottom layers are also exchanged. Now, the top layer of CGL is made of HAT-CN and bottom layer is made of TAPC. Because of this, electrons in top layer starts repelling by cathode and move to the emissive layer. In the same manner, holes in bottom layer will be repelled by anode and reach to the emissive layer. So, electron and hole concentrations are increased inside the emissive layer as this layer already contains charge carriers which have generated from cathode and anode. Hence, according to the Langevin's recombination rate, increment in charge carriers will directly improve the recombination rate which significantly enhanced the performance of D₆ in terms of current and luminescence. The total thickness of CGL in D₆ is considered as 40nm due to some structural variations as shown in figure below.

Figure 7 is showing the device structure of D₆ having the thickness and materials. The maximum current and luminescence of proposed device is obtained as 0.44 A and 3636.3 cd/m² as highlighted in Fig. 8. The device exhibits excellent luminescence which is about 16.8, 2.3, 1.7, 3 and 1.6 times better as compared with D₁, D₂, D₃, D₄ and D₅, correspondingly.

Thickness Optimization: In this section, the thickness of charge generation layer in proposed device (D₆) is reduced to achieve an optimized device. This process is done in three ways which are: 1) reduction of thickness in both layers of CGL evenly, 2) fix the thickness of HAT-CN layer at 20nm and reduce the thickness of TAPC layer and 3) fix the thickness of TAPC layer at 20nm and reduce the thickness of HAT-CN layer. The thickness reduction is performed by reducing 2nm thickness in every step as shown in Fig. 9, Fig. 10, and Fig. 11. All the three cases are discussed in details below.

(a) Case I: Initially, the total thickness of CGL is considered as 40nm. As shown in Fig. 9 (a), in Case I both layers of CGL are decreased five times by 2nm of thickness each i.e. layer of TAPC by 2nm and layer of HAT-CN by 2nm (total variation of 4nm). Eventually, the total thickness of CGL is reduced upto 24nm from 40nm which provides about 1.3 times better luminescence. In this process, the minimum and maximum
luminescence is achieved as 3640 cd/m$^2$ and 4670 cd/m$^2$ at 40nm and 24nm thickness of CGL, correspondingly. This data can be clearly verified from Fig. 9 (b) as it has better clarity.

(b) Case II: In this case, only layer of TAPC is varied from 20nm to 12nm whereas the layer of HAT-CN material is kept fixed at 20nm. Here, the curve for luminescence is drawn at 20nm, 18nm, 16nm, 14nm and 12nm thicknesses of layer of TAPC material as shown in Fig. 10 (a). The maximum luminescence is achieved as 4080 cd/m$^2$. This small change can be noticed in Fig. 9 (b) which is a zoomed version of Fig. 10 (a).

(c) Case III: This case is just reverse of case II. The thickness of HAT-CN material is varied from 20nm to 12nm in five steps and TAPC is constant at 20nm as depicted in Fig. 11 (a). The peak luminescence is obtained as 4150 cd/m$^2$. Here, Fig. 11 (b) is a zoomed view of Fig. 11 (a) where small variation can be seen easily.

<table>
<thead>
<tr>
<th>Case</th>
<th>Thickness (nm)</th>
<th>Luminescence (cd/m$^2$)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>TAPC</td>
<td>HAT-CN</td>
</tr>
<tr>
<td>I</td>
<td>20</td>
<td>20</td>
</tr>
<tr>
<td></td>
<td>18</td>
<td>18</td>
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<tr>
<td></td>
<td>16</td>
<td>16</td>
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<tr>
<td>II</td>
<td>18</td>
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<td>III</td>
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<td>14</td>
<td></td>
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<td></td>
<td>12</td>
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</table>
In all the three cases discussed above, the maximum luminescence is achieved in Case I i.e. 4670 cd/m$^2$ at 24nm thickness of CGL as highlighted in Table 3. This value is almost 78% improved as compared with the device ($D_6$) where thickness of CGL is 40nm. Also, in Case II and III, Case III is exhibiting better results. In case I, both the top and bottom layers are varied in the same ratio of 2nm. So, electrons and holes will travel fast towards the emissive layer from both sides as both layers are equally closer to the emissive layer. On the contrary, in Case II and Case III, electrons and holes, respectively will take more time on comparing with Case I. This will directly affect the recombination of charge carriers. Hence the peak luminescence values of both cases are less as compared with Case I. Moreover, if Case II and Case III are compared, in Case III the layer of TAPC in fixed which contains holes. And it is known that in organic electronics the mobility of holes is better than electrons so hole can travel fast to the emissive layer [28]. This provides better luminous properties to Case III as recombination will be higher in this case.

5. Conclusion

Basically, six different blue OLEDs $D_1$, $D_2$, $D_3$, $D_4$, $D_5$ and $D_6$ are compared in this paper. Here device $D_1$ is without CGL and other five devices including the proposed device contain CGL at different positions. The CGL comprises of two different layers in which top layer behaves as hole injector i.e. 1,1-bis[(di-4-tolyamino)phenyl)]cyclohexane (TAPC) and bottom layer acts as electron injector i.e. 1,4,5,8,9,11-hexaaazatriphenylene-hexacarbonitrile (HAT-CN). The combination of these two materials provides easy transfer of charge carriers into the emissive layer. Hence, devices in which CGL is incorporated provide higher current and excellent luminescence. In this paper, the proposed device where CGL is utilized outside of the emissive layer showed significant improvement as the parameters such as luminescence and current are reported as 0.44 A and 3636.3 cd/m$^2$, respectively. Addition to this, the thickness optimization of the proposed device is also incorporated by the varying the thickness of utilized CGL layer.

Declarations

Consent for publication

All the authors understand and give their consent for the publication of manuscript “Characteristics performance and analysis of positional variation in charge generation layer to enhance performance of OLED” in “Journal of Computational Electronics” Journal.

Availability of data and materials

Not Applicable

Competing interests
No, all the authors declare that they have no competing interests as defined by Springer, or other interests that might be perceived to influence the results and/or discussion reported in this paper. The authors have no conflicts of interest to declare that are relevant to the content of this article.

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**Author's Contributions**

**Sugandha Yadav:** Conceptualization, Methodology, Data curation, Writing – original draft, Writing – review & editing.

**Poornima Mittal:** Conceptualization, Methodology, Supervision, Data curation, Writing – review & editing, Validation.

**Shubham Negi:** Supervision, Writing – review & editing, Visualization, Investigation, Formal analysis.

**Availability of data and materials**

Data and materials utilized and supporting the results of this study are available within the article.

**References**


22. Y. Liu, X. Wu, Z. Xiao, J. Gao, J. Zhang, H. Rui, X. Lin, N. Zhang, Y. Hua and S. Yin, Highly efficient tandem OLED based on C60/rubrene: MoO3 as charge generation layer and LiF/Al as electron


Figures
Figure 1

Structure of Blue OLED (D₁) [26]

Figure 2

Device structure of D₁ along with the thickness and materials

Figure 3

(a) Reference and experimental data of current & (b) Combined curve for current density and luminescence of device D₁
Figure 4

Blue OLED with (a) CGL below cathode (D_2), (b) CGL below anode (D_3), (c) CGL at both sides of electrodes (D_4), and (d) CGL inside emissive layer (D_5)
Figure 5

(a) Current curve and (b) Luminescence curve for all devices (D₁ - D₅)

Figure 6

Proposed Device (D₆)
Figure 7
Device structure of $D_6$ along with their thickness and materials

Figure 8
(a) Current and luminescence curves of proposed device ($D_6$) and (b) Luminescence comparison of $D_1$, $D_5$ and $D_6$
Figure 9

(a) Luminescence comparison at different thickness of top and bottom layer of CGL and (b) the zoomed view of curve (a) to show the variations in luminescence.

Figure 10

(a) Luminescence comparison at different thickness of bottom layer where top layer is fixed of CGL and (b) the zoomed view of curve (a) to outline the variations in luminescence.
Figure 11

(a) Luminescence comparison at different thickness of top layer where bottom layer is fixed of CGL and (b) the zoomed view of curve (a) to depict the variations in luminescence