International Journal Of Electronics Engineering (Issn: 0973-7383)
Volume 10 • Issue 2 pp. 329-335 June 2018-Dec 2018 www.csjournals.com

Performance Enhancement of Tandem Light Emitting Diode Using Organic Interconnector

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Abstract: The planar organic charge generation layer (CGL) of C70 (Fullerene)/ CoPc (Cobalt Phthalocyanine) based highly efficient red tandem device is proposed in this paper. It is found that there is large energy alignment of CoPc with the neighbouring electron transport layer thereby reducing the energy barrier for the charge carriers injection. The organic hetero junction used generate charges as well as exhibit organic photovoltaic behaviour as the absorption spectra of charge generation layer coincides with the photo luminescent spectra of emitter in the red wavelength zone i.e from 600-800 nm used thereby absorbing the red wasted photons and thus enhancing the lifetime of the device. In addition an ultra thin layer of Al/LiF is used as electrons injection layer (EIL) so as to enhance the injection of electrons from CGL to emissive unit. It is found that the tandem red device can reach current efficiency and luminance of 26 cd/A and 26039 cd/ m^2 at 20 mA/ cm^2 and 100 mA/ cm^2 respectively which is 1.663 times and 1.665 times higher than the corresponding single emitter device. Thus, the effective charge generation and OPV function of proposed CGL as well as remarkably high injection property of electron injection layer leads to the high performance tandem OLED.

Keywords: Charge Generation Layer; Efficiency Improvement; Electron Injection; Tandem OLED.

Introduction

Tandem OLEDs are the extension of conventional OLED structure. The concept of Tandem OLEDs was first launched in the year 2003 and since then they have grabbed the eyeballs due to their tendency to produce higher efficiency in contrast to conventional OLEDs. They produce relatively high luminance at minuscule current density than the conventional OLEDs thereby reducing the efficiency roll off caused by higher current density in addition to the elongated lifespan in contrast to conventional OLEDs. They consist of more than one electroluminescent units that are linked electrically in series through charge generation unit which serves as heart of tandem OLEDs. The electroluminescent unit is a multi layer structure consisting of hole transport layer, emitting layer, electron transport layer all contained in between anode and cathode respectively. The charge generation unit is basically a bilayer architecture or most commonly a PN junction that comprises of both p-type and n-type organic layers so as to allow the charge carriers generation as well their transport [9]-[1]-[3]. The charge generation layer generates as well as inject the charges into the neighboring emitting on application of the external supply for the effective charge balance in the emitting unit. One of the most important aspect for developing high performance applications is that the mobility of charge carriers should be very high and for this the charge generation layer as well the transport and the injection layers should be highly transparent and should offer less resistance.

When the external voltage is supplied, positive and negative charge carriers are injected by anode and cathode respectively. At the same time, charges carriers in pairs are produced at the organic hetero junction. These charge carriers move to the emitting layer i.e. electrons from the lowest unoccupy molecule orbit level of n-type layer travels to the lowest unoccupy molecule orbit level of adjacent electron transport and injection layer from where they move to the lowest unoccupy molecule orbit level of the emitting layer where they reunion with holes injected by the anode at the highest occupy molecule orbit level of the emitting layer to produce and emit. At the same time, holes from the highest occupy molecule orbit level of p-type layer travels to the highest occupy molecule orbit level of adjacent hole transport and injection layer from where they move to the highest occupy molecule orbit level of the emitting layer and reunite with electrons injected by the cathode at the lowest unoccupy molecule orbit level of the emitting layer to produce and emit photons. Thus light output gets enhanced with the use of more number of electroluminescent units[4]-[10]. The performance of the tandem device increases with the increase in number of electroluminescent units.

The enhanced performance of Tandem OLED is attributed to the charge generation layer which serves exactly like the heart of Tandem OLEDs. For the large displays fabrication, tandem devices are more suitable as in these devices, there are more layers between the emitting unit and electrodes, thus any types of shorts can be avoided. In tandem devices, it is possible to build different color combinations because of the presence of two or more emitters thus producing more vibrant colors. As a fundamental interconnector in Tandem

OLEDs, CGLs must have high transparency, low electrical resistance, matched HOMO LUMO levels with the adjacent transport layers, low power loss etc. When the external voltage is supplied, charge carriers are generated in the CGL, which are then injected into the neighboring emitting unit via the transport layers, where they combine with the opposite charges injected by the electrodes to generate light [11]-[14]. The CGL is typically a p-n junction that, consists of metal/metal or metal oxide/metal oxide bilayer structure, organic n-type layer/p-type layer or organic photovoltaic heterojunctions. However, CGLs having metal layers (metal oxide layers) have many limitations like low transmission capability and moreover, some of the metal oxides used in CGLs can react with the negatively charged dopants present in the electron transport layer thereby the doubling the driving voltage and reducing the performance of the device. Also, out of the total generated light in the emissive layer, only 20% of the light can be extracted out of OLED, the rest 80% of the remaining light is lost because of optical losses and surface Plasmon or waveguide modes.

Therefore, it becomes very crucial to employ that lost part of light for the enhanced EL emission and efficiency of device. To overcome all these problems organic photovoltaic (OPV) type charge generation units are generally employed. [15]-[19].They have the tendency to generate the charges called electrically generated charges upon the application of external voltage as well as they have the additional advantage of absorbing a fraction of wasted photons (packets of light energy) from the emissive layer to form the excitons and then at the interface of heterojunction separates them into free electrons and holes. These free electrons and holes are known as photo generated charges. Thus, due to their unique feature of producing photo as well as electric generated charges, this OPV heterojunctions are contemplated as very efficient interconnectors in tandem OLEDs. In this work, the conjunction of organic fullerene derivative C70 and CoPc (Cobalt Phthalocyanine) heterojunction are proposed as planar charge generation layer of tandem OLEDs, where C70 behave as n-type semiconductor material that has high thermal stability with exceptionally high electrons mobility and CoPc act as p-type semiconductor material that has high holes mobility. Further, to boost up the injections of electrons from the CGL to the neighboring emitting unit, the combination of thin layers of Al (aluminium) along with LiF (Lithium Fluoride) is used as electron injection material. The advantage of using aluminium is that it helps in decreasing the work function of C70, thus increasing the device performance.

Materials and Methods

The structure adopted for Al/LiF/ C70/ Cobalt pthalocyanine (CoPc) charge generation layer based tandem device is a planar heterojunction architecture as shown in figure 3.2 with the layers configuration of Anode of highly conductive and transparent Indium Tin Oxide (ITO)/ HIL Molybdenum Trioxide (MoO_3)/ HTL N,N0-bis-(3-naphthy)-N,N'-biphenyl-(1,1'-biphenyl)-4,4'-diamine (NPB)/ red emitting layer N,N'-Bis(naphthalen-1-yl)-N,N'-bis(phenyl)-2,2-diMe (α-NPD): Tris [1-phenylisoquinoline-C^2,N] iridium(III ($Ir(piq)_3$)/ ETL 4,7-diphenyl-1,10-phenanthroline (Bphen) /coalition of Lithium Fluoride (LiF) and Aluminium (Al) as cathode. The CGL comprises the multi-layers of LiF/ Al/ Fullerene Derivative (C60)/ CoPc in which C70 act as n-type semiconductor material that is an excellent thermally stable material and has excellent electrons mobility and Pentacene act as the p-type semiconductor material with high holes mobility. Further, to boost up the injections of electrons from the CGL to the neighbouring emitting unit, the coalition of thin layers of Al along with LiF is used as n-type CGL supporting layers. The detail layer structure of red single emitting and proposed Tandem device is summarized in Table 1. To prove the efficacy of proposed OPV planar heterojunction and the ultra thin electron injection layer (Al/LiF), the conventional single emitter device structure is kept similar as used in ref. [15] except that of the emitter that was CBP: $Ir(piq)_2$acac (8% by volume). Due to the simulations environment limitation, the emitter used in this work is α-NPD: $Ir(piq)_3$ (20% by volume). However, the emitting zone and the peak wavelength of α-NPD: $Ir(piq)_3$ is same as that CBP: $Ir(piq)_2$ acac. Both the emitters emit in the red wavelength zone of 600-800 nm and has peak wavelength around 625nm. The charge generation layer used in ref. [15] comprised of multi-layers of LiF/C60/CuPc/ MoO_3, where C60/CuPc exhibits the organic photovoltaic type behavior and LiF and MoO_3 are the interface layers that increases the injection of charge carriers from CGL to the emissive layer.

Table 1: The detail device structures of single emitting and red Tandem OLED

Device	Device Name	Layer Structure
Red conventional OLED	A	ITO/ EL/ LiF (2 nm)/Al (150 nm)
Red Tandem OLED	R	ITO/ EL/ CGL/ EL /LiF (2 nm)/Al (150 nm)

EL: MoO_3 (3 nm)/ NPB (40 nm)/α-NPD: $Ir(piq)_3$ (30 nm, 20% by volume)/Bphen (30 nm)
CGL: Al (3 nm)/ LiF (2 nm)/ C70 (5 nm)/ CoPc (5 nm)

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Results and Discussions

The main purpose of Charge generation layer is to produce charges. The detail layer structure of tandem device using the proposed charge generation layer is shown in Fig. 1. The device simulations are performed using Fluxim SETFOS 4.5 software.The energy level alignment of CGL with the adjacent transport layers is depicted in Fig. 2. It can be seen that the HOMO **(**Highest occupied molecular orbit) level difference between NPB (5.4 eV) and CoPc (5.2eV) is only 0.2eV and LUMO (Lowest unoccupied molecular orbit) level difference between C70 (3.9 eV) and Bphen (3.1 eV) is 0.8 eV. Due to the small energy level difference between CGL and the neighboring transport layers, free charge carriers are produced at the interface of C70/CoPc planar heterojunction, i.e. holes can easily transfer from HOMO of CoPc to that of NPB while electrons can easily jump from LUMO of C70 to that of Bphen. Then from HOMO of NPB and LUMO of Bphen, the holes and electrons can move easily to the adjacent emitting units.

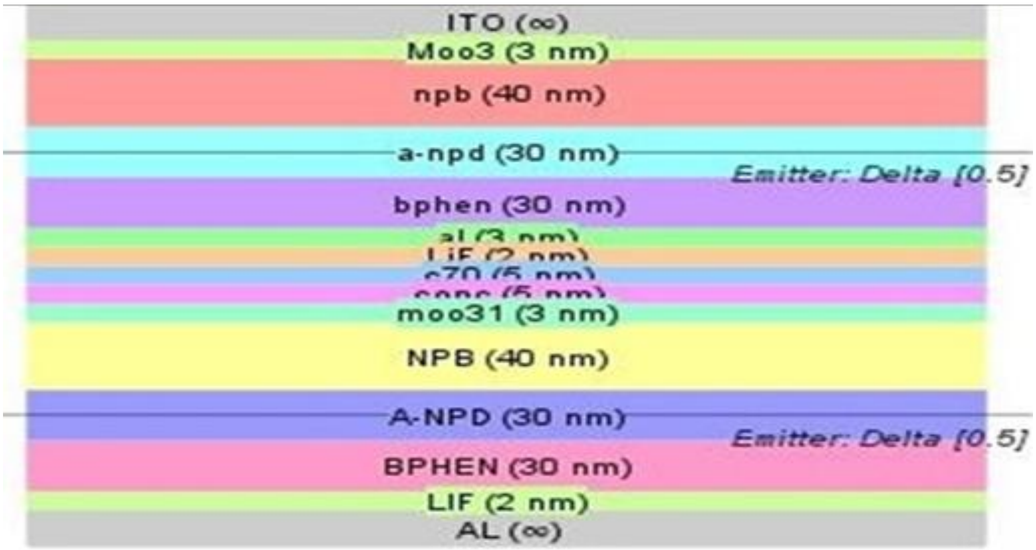

Figure 1. Proposed Device Structure

The optical and electrical properties are used to study the effect of thickness variation of CGL on the behavior of the device. The effect of thickness vof CGL on the optical properties is studied by calculating the transmittance for different thickness of C70/CoPc from the absorption spectra as shown in Fig. 3 by using the formula Transmittance= antilog (2-absorption). Thus, transmittance is calculated to be 94.5% from the absorbance graph and using above stated formula.Thus CGL is highly transparent.

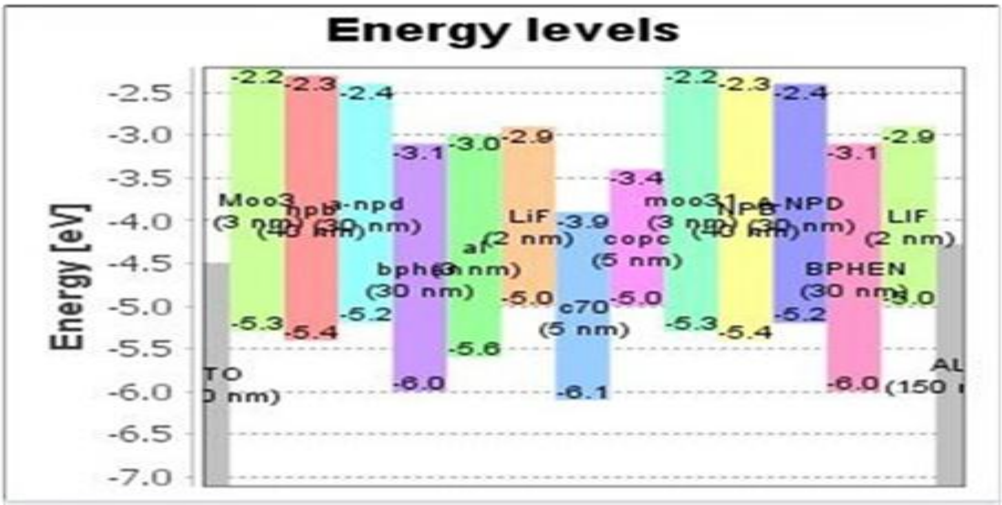

Figure 2. Energy level diagram of various layers used in proposed design

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Volume 10 • Issue 2 pp. 329-335 June 2018-Dec 2018 www.csjournals.com

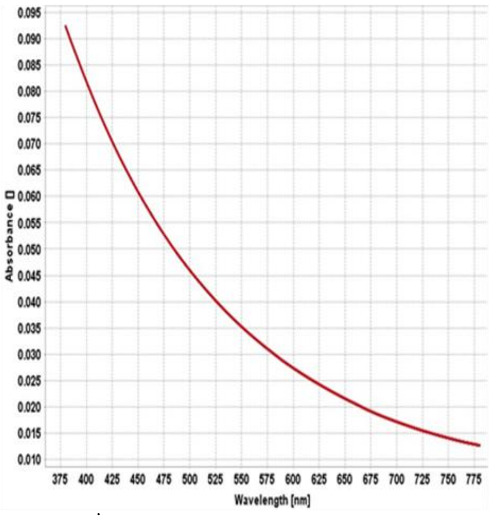

Figure 3. Absorbance plot of CGL at 5 nm thickness

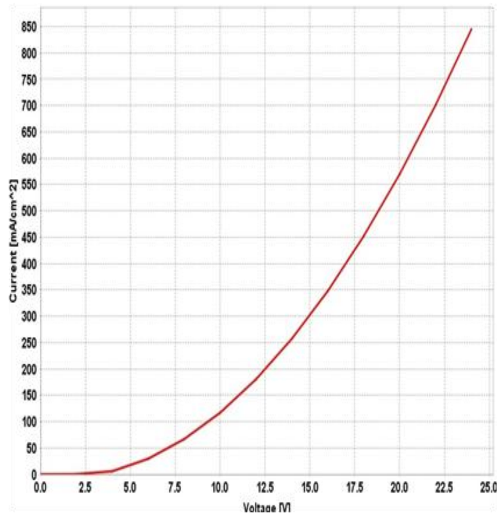

Figure 4. Current Density plot of CGL at 5 nm thickness

The effect of thickness variation of CGL on the electrical properties is studied by observing the current density versus voltage plot of different thickness of C70/CoPc. A test device T with the structure of ITO/Bphen (30 nm)/ Al (3 nm)/ LiF (2 nm)/ C70 (5 nm)/ CoPc (5 nm)/ NPB (40 nm)/ Al is designed and simulated for this purpose. The voltages at 200 mA/cm^2 is noted From Fig. 4 it is found that voltages is 12.45 V for thickness (5nm, 5nm). Thus the small voltage is found for 5nm thickness of both C70 and CoPc for the same current density. Thus, it can be noted that increasing the thickness of CGL causes the difficulty in electron extraction from CGL. That is why, more voltage is required to extract the electrons at same current density with the increasing thickness [20]-[21]. Therefore, the optimized thickness of both C70 and CoPc is found to be 5nm from both the electrical and optical properties.

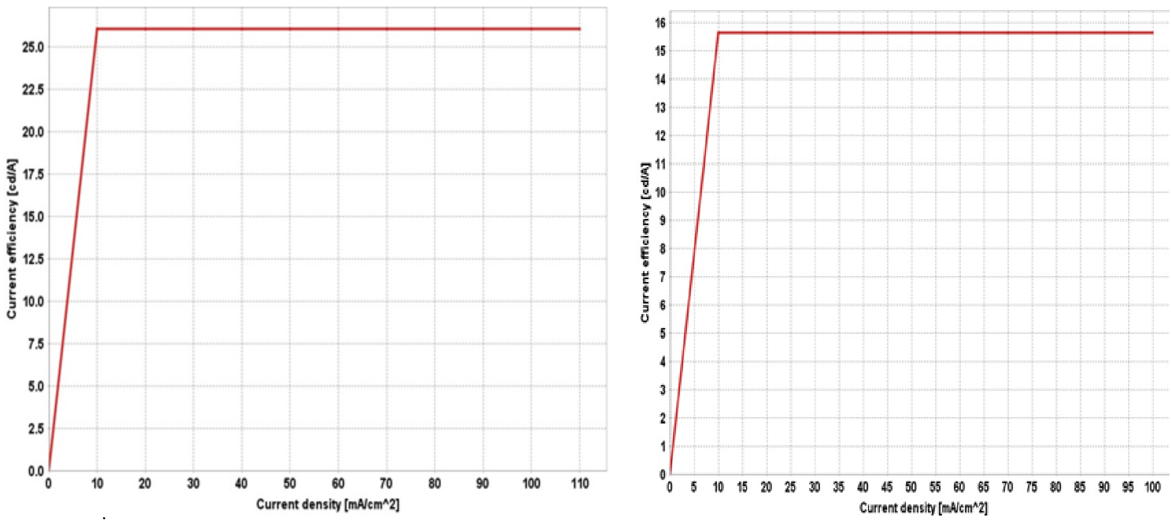

Figure 5. Current Efficiency plot of Device R, Device A

Further, tandem devices is designed and simulated to prove the effectiveness of proposed CGL consisting of multi layers of Al/LiF/C70/CoPc.. It can be shown from the current efficiency as given in Fig. 5a, Fig. 5b and luminance plots of red tandem device R and device A as given in Fig. 6a,Fig. 6b respectively that the red tandem device R with CGL layer consisting of Al/LiF/C70/CoPc shows the best performance in terms of current efficiency and luminance with respect to single emitter device A. The current efficiency of device is found to be 26 cd/A at 20 mA/cm^2 and luminance is 26039 cd/m^2 at 100 mA/cm^2. It can be observed that the device R exhibits almost two times enhanced current efficiency and luminance in contrast to device A (15.63 cd/A and 15,630 cd/m^2).

This enhanced performance can be attributed to the interface layer or injection layer used with CGL which are crucial because of their functional efficacy and excellent charge injection property which eventually causes carrier recombination and balance in the emitting layer and also there is a large energy level alignment between CoPc and C70 as well as good transport ability of C70 than that of C60. The HOMO of CoPc is 5.2 eV and LUMO of C60 is 3.7 eV whereas that of C70 is 3.9 eV. Therefore electrons have to cross a large energy barrier of 1.5 eV to jump to LUMO of C60 from the HOMO of CoPc in contrast to C70/CoPc where the energy barrier is 1.3 eV. Thus C70/CoPc heterojunction is highly efficient than the C60/CoPc heterojunction [22]. Further enhancement in EQE of device R is also observed in contrast to device A. The detail EL performances of the device R and A is concluded in Table 2.

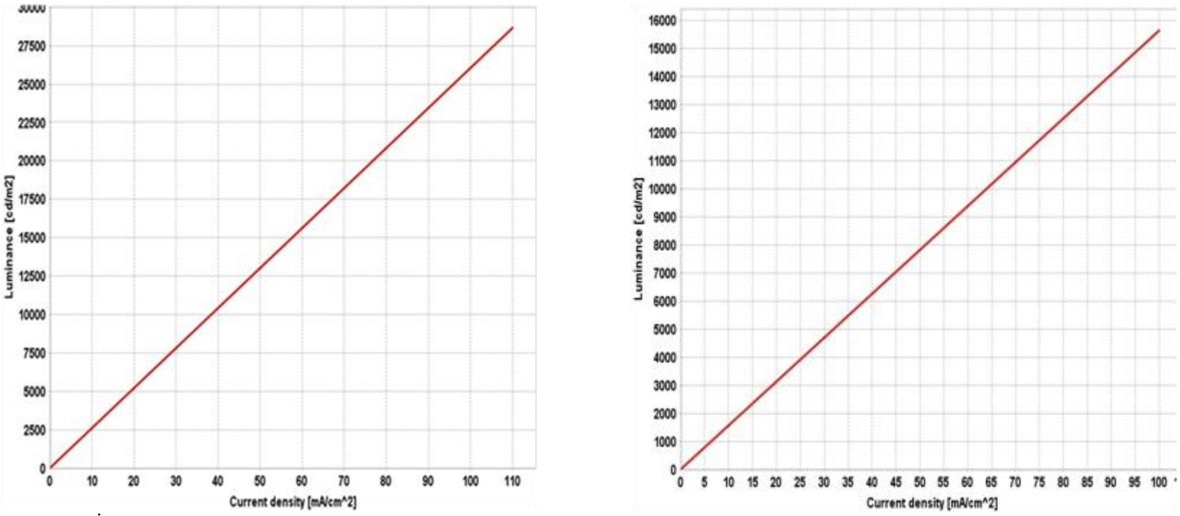

Figure 6. Luminance plot of Device R, Device A

From current density versus voltage plot studied at 1000 cd/m^2 (Fig. 6) it is found that current density of structure A is 6.39 mA/ cm^2 which is 1.6 times higher than device R having current density of 3.84 mA/ cm^2. Thus device R requires less current density than the device A to reach the same luminance. Therefore there is slow efficiency roll off in device R than device A which increases with the current density. Thus, Tandem devices are more efficient than conventional devices.

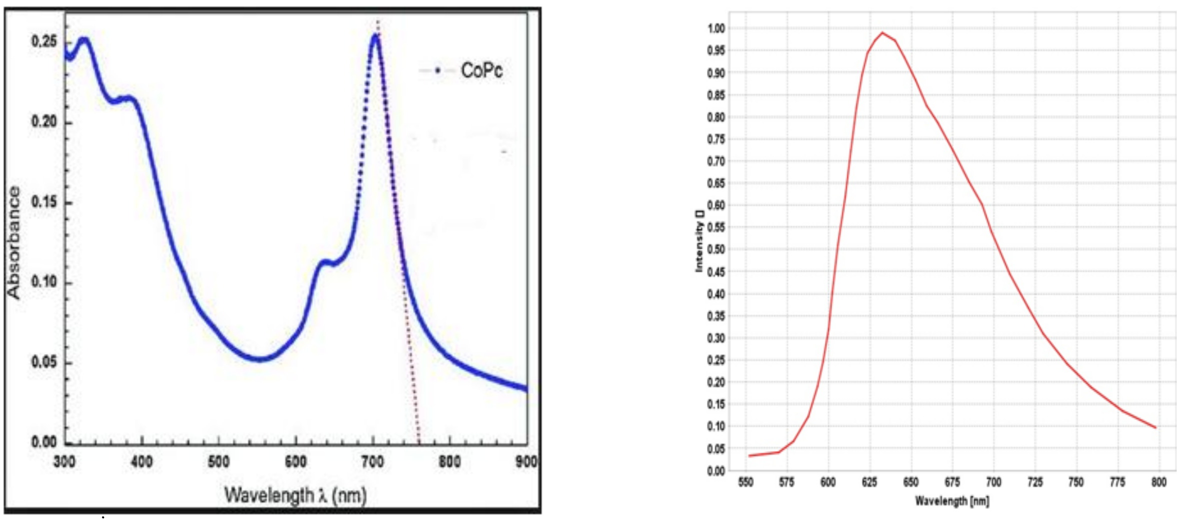

Figure 7. Absorption spectra of CGL[23], PL spectra of red emitter

The difference in the performance of tandem device and single emitter unit device is because the charge carriers generated at the CGL exist in pairs always, making the more recombinations in the emitting unit thus leading to more charge balance in contrast to conventional OLEDs.. The enhancement in performance of tandem devices is because of the CGL that produced almost the same number of free charges as are inoculated by the anode and cathode thus causing charge carriers balance which leads to reduction in quenching and the plasmonic losses and also increases the microcavity effect. In tandem devices, there are more than one electroluminescent unit, hence the emitting unit that are more close to the metallic electrode will only get affected by the plasmonic quenching of the electrode, rest will be saved, thus leading to more light outcoupling. For the large displays fabrication, tandem devices are more suitable as in these devices, there are more layers between the emitting unit and electrodes, thus any types of shorts can be avoided.

To observe the OPV behavior of proposed heterojunction, the UV vis absorption spectra of C70/CoPc and the photoluminescent spectra of α-NPD: $Ir(piq)_3$ is studied. As shown in Fig. 7 both the spectra overlap each other in the red emitter zone of 600-800 nm. So it can be presumed that the red wasted photons due to surface plasmon and waveguide modes in the emitter can be easily absorbed by C70/CoPc heterojunction to form excitons and further dissociate them into free charges thus enhancing the efiiciency.

.The basic requirement in CGL is the generation of charges at the CGL interface. As known and proved in that the charges are produced at C70/CoPc heterojunction because of the transfer of electrons from CoPc to C70. The entire working process can be concluded as follows. At small current density, due to the generation of charges at heterojunction, current efficiency increases rapidly with the increase in luminance of device. Then, the current efficiency becomes invariable, after attaining the maximum height. This is because of the recombination of these generated charges at CGL with the charges inoculated by electrodes in the emissive layer to produce photons. Due to the OPV type behavior, CGL fetch a part of wasted photons from emissive layer to generate more charges and again the whole process repeats causing the slower efficiency roll off. Thus, this OPV type CGL is very effective and results in longer lifespan of the device.

Table 2. Comparison of proposed devices with the corresponding devices used in Ref. [15]

Parameters	Red Single Emitter Device A	Red Single Emitter Device [15]	Red Tandem Device R	Red Tandem Device [15]
Current Efficiency (cd/A) @ 20 mA/ cm^2	15.63	8	26	16
Luminance (cd/m^2) @ 100 mA/ cm^2	15630	7593	26039	16061
EQE @ 1000 cd/ m^2	17.9	8.8	29.3	14.1

Acknowledgment
The author would like to thank Dr. S.S Pattnaik, Director National Institute of Technical Teachers' Training & Research, Chandigarh, India and for constant motivation and support throughout this research work.

Conclusion

The coalition of C70/CoPc along with the effective electron injection layer comprising the combination of aluminum and lithium fluoride is proposed as planar organic heterojunction. The absorption and PL spectra of both CGL and red emitter coincide in red wavelength range from 600-800 nm showing organic photovoltaic behavior, so the proposed CGL can absorb red wasted photons from red emitter. Thus, efficiency roll off gets reduced in contrast to devices using other types of charge generation layers. Also, there is a large improvement in current efficiency and luminance of red tandem device R (26 cd/A and 26039 cd/m^2 at 20 mA/cm^2 and 100 mA/cm^2 respectively) in contrast to single emitter device A (15.63 cd/A and 15,630 cd/m^2). The improvement in performance of device R is due to the OPV function of CGL and efficient electron injection capability of thin Al/LiF and also the large energy alignment of CoPc with the neighboring electron transport layer thereby reducing the energy barrier for the charge carriers injection. Thus, the doping free planar organic heterojunction yield high performance tandem OLED with the low fabrication cost.

References

[1] Y. Karzazi, “Organic Light Emitting Diodes: Devices and Applications,” J. Mat. Env. Sci., vol. 5, no. 1, pp. 1-12, January, 2014.

[2] N.Thejo. Kalyani, S.J. Dhoble, “Organic Light Emitting Diodes: Energy Saving LightingTechnology A Review,” J. Renew. Sust. Energy Rev., vol. 16, no. 5, pp. 2696– 2723, March 2012.

[3] T. D. Schmidt, B. J. Scholz, C. Mayr and W. Brutting, “Efficiency Analysis of Organic Light-Emitting Diodes Based on Optical Simulations”, J. Sel. Top. Quant. Elect., vol. 19, no. 5, pp. 1-12, October 2013.

[4] Z. Ma, S.Q. Liu, S. Hu and J. Yu, “Highly efficient tandem organic light-emitting diodes based on SubPc: C60 bulk heterojunction as charge generation layer”, J. Lum., vol. 39, pp. 29–34, January 2016.

[5] J. Liu, X. Shi, X. Wu, J. Wang and G. He, “Inverted Tandem Phosphorescence Organic Light-Emitting Diodes Based on MoO3/Al/Cs2CO3 Charge Generation Unit,” J. Disp. Tech, vol. 11, no. 4, pp. 341-345, January 2015.

[6] F. A. Angel, J. U. Wallace and W. Tang, “Effect of lithium and silver diffusion in single-stack and tandem OLED devices,” J. Org. Elect., vol. 42, pp. 102-106, December 2016.

[7] Z. Lu, X. Li, Y. Wang, J. Xiao and P. Xu, “Field- and thickness dependent performance of tandem organic light-emitting diodes with buffer-modified fullerene/copper phthalocyanine heterojunction as interconnector,” J. Curr. Appl. Phys., vol. 14, pp. 1465-1469, November 2014.

[8] J.P. Yang, W.Q. Wang, L.T. Shang, et al., “Dynamic processes of charges generation in intermediate connectors for tandem organic light emitting diodes,” J. Org. Elect., vol. 46, pp. 145-149, April 2017.

[9] L. Li, M. Guan, G. Cao, Y. Li and Y. Zeng, “Tandem organic light-emitting diodes with an effective charge-generation connection structure,” J. Short State Comm., vol. 150, no. 35-36, pp. 83-85, June 2010.

[10] Q. Guo, H. Sun, J. Wang, D. Yang, J. Chen and D. Ma, “Charge generation mechanism of tandem organic light emitting diodes with pentacene/C70 organic heterojunction as the connecting layer,” J. Mat. Chem. vol. 4, no. 2, pp. 1-9, January 2013.

[11] Y. B. Zhao, S.T. Tan, H.V. Demir and X. W. Sun, “Highly stable and high power efficiency tandem organic light-emitting diodes with transition metal oxide-based charge generation layers,” J. Org. Elect., vol. 23, pp. 70-75, August 2015.

[12] H. Zhang, Y. Dai, D. Ma, and H. Zhang, “High efficiency tandem organic light emitting devices with Al/WO3/Au interconnecting layer,” Appl. Phys. Lett., Vol. 91, No. 12, pp. 1-3, September 2007.

[13] H. Yang, J. Kim, K. Yamamoto and H. Hosono, “Efficient charge generation layer for tandem OLEDs: Bi-layered MoO3/ZnO-based oxide semiconductor,” J. Org. Elect., vol. 46, pp. 133-138, March 2017.

[14] Z. Ma, J. Zhao, X. Wang and J. Yu, “Effect of bulk and planar heterojunctions based charge generation layers on the performance of tandem organic light-emitting diodes,” J. Org. Elect., vol. 30, pp. 136-142, March 2016.

[15] D. Zhao, H. Liu, Y. Miao, et al., “A red tandem organic light-emitting diode based on organic photovoltaic type charge generation layer,” J. Org. Elect., vol. 32, pp. 1-6, May 2016.

[16] Z. Lua, Y. Wang, X. Li, J. Xiao and Z. Deng, “Role of MoO3-modified organic photovoltaic-type charge generation layer in tandem organic light-emitting Diodes,” J. Syn. Met., vol. 229, pp. 47-51, July 2017.

[17] M. Ramar, P. Tyagi, C. K. Suman and R. Srivastava, “Enhanced carrier transport in tris(8 hydroxyquinolinate) aluminum by titanyl phthalocyanine doping,” J. Roy. Soc. Chem. Adv., vol. 4, pp. 56-61, September 2014.

[18] S. Altazin, S. Züfle, E. Knapp, C. Kirsch, et al., “Simulation of OLEDs with a polar electron transport layer,” J. Org. Elect., vol. 39, pp. 44-49, December 2016.

[19] J.P. Yang, Y. Xiao, Y.H. Deng, et al., “Electric-Field-Assisted Charge Generation and Separation Process in Transition Metal Oxide-Based Interconnectors for Tandem Organic Light-Emitting Diodes,” J. Adv. Funct. Mat., vol. 22, no. 3, pp. 600-608, November 2011.

[20] Y.H. Chen, Q. Wang, J.S. Chen, D.G. Ma, D.H. Yan and L.X. Wang, “Organic semiconductor heterojunction as charge generation layer in tandem organic light emitting diodes for high power efficiency,” J. Org. Elect., vol. 13, pp. 21-28, April 2012.

[21] Y.H. Chen, D.G. Ma, H.D. Sun, et al., “Organic semiconductor heterojunctions: electrode-independent charge injectors for high-performance organic light-emitting diodes,” J. Light Sci. Appl., vol. 5, pp. 1-7, March 2016.

[22] L. Duan, T. Tsuboi, Y. Qiu, Y. Li and G. Zhang, “Tandem organic light-emitting diodes with KBH4 doped 9,10-bis(3-(pyridine-3-yl)phenyl) anthracene connected to the charge generation layer,” J. Opt. Exp., vol. 20, no. 3, pp. 64-72, June 2012..

[23] Ahmet T.Bilgiçli, Meryem N. Yaraşır, Mehmet Kandaz, Cihat Ilik, Ahmet Demir, Sadik Bagcik, “Non peripheral tetra phthalocyanines bearing alkyl chain moiety; Synthesis, characterization and fabrication of the OFET based on phthalocyanine, Synthetic Metals, vol. 206, pp. 33-41, May 2015.