Int. J. Thin.Film. Sci. Tec. **11**, No. 2, 207-211 (2022)

International Journal of Thin Films Science and Technology

http://dx.doi.org/10.18576/ijtfst/110208

Impact of Different Anode Materials on Performance of Organic Light Emitting Diodes

Aditya Tiwari[*] *and Brijesh Kumar*

Department of Electronics & Communication Engineering, Madan Mohan Malaviya University of Technology, Gorakhpur, India.

Received: 20 Feb. 2022, Revised: 1 Apr. 2022, Accepted: 22 Apr. 2022

Abstract: Organic light-emitting diodes (OLEDs) are one of the most explored organic electronic devices. OLEDs are flexible, light-weight, have deep blacks and better contrast ratio when compared to other display technologies, therefore, they are considered to be the "bleeding edge" in display technology, wherein every flagship display device OLEDs are being used in place of LCDs. This paper essentially portrays how the anode material affects the overall device performance in OLEDs. Recent historical surveys have depicted that altering the work-function of the anode will have a huge impact on various performance parameters of the device. In this paper, we take six experimental device architectures, all similar but with different anode materials. The goal here is to enhance device performance. Different anode materials like graphene, PEDOT: PSS, PEDOT: PSS/GO composite are mainly compared with traditionally used ITO based anodes. An overall increment in luminance power, current density, electron-hole concentration, and exciton density is seen. PEDOT: PSS/GO composite anode also shows an enhanced performance of about 45% over ITO based OLED devices. It can also be used in various optoelectronic devices, owing to its transparency and smooth working operation. Further analysis has been done to examine the plots and graphs to see the viability of other experimental devices too.

Keywords: OLED, Anode, ITO, Graphene, and PEDOT: PSS.

1 Introduction

Organic light emitting diode or OLED is one of the most dynamic field of technology. It has attracted huge amount of interest in its usage in display devices. This is due to the fact that it has great benefits, which are an easy fabrication method, a fast switching speed, a wider viewing angle and also a lower fabrication cost when compared with an orthodox Liquid Crystal Display (LCD) [1,2,3,4].

Due to the large scale arduous effort from both scientific and engineering community, the field of organic electronics have experienced rapid development in recent times. Enormous efforts are been made in order to increase the performance of OLED either by altering the interface structure or by the use of different materials for making various layers of the device for achieving better overall injection of charge carriers [5]. These facts are the reasons due to which OLEDs are the most advance field of organic electronics.

Now, to augment the performance of OLEDs, it is fundamental to create and develop a better optimized device structural layout. The main goal for improving the present device efficiency should be the enhancement of injection of charge carriers from anode and cathode into various intermediate layers and finally to recombination layer.

In the extensive research which is being carried out lately by the research community it is noted that the properties of anode, particularly the anode and the hole transport layer (HTL) interface is very vital in enhancing the efficiency, performance, and longevity of organic light emitting diodes (OLEDs) [5,6,7]. Defects in the anode surface reduces the anode-organic film interface adhesion, which then increases the electrical resistance, and this results in the development of non-emissive dark spots in the OLED material, which greatly affect the life-cycle of OLED devices [6]. This makes the selection of anode material very important in the topography of OLEDs. In the past several compounds have been used for anode material with the likes of indium tin oxide (ITO), graphene, PEDOT-PSS, PEDOT-PSS/GO composite, carbon Nano-tubes etc. This paper mainly focuses on a comparative study of various materials as anode in OLEDs.

Indium tin oxide (ITO) is a natively used anode material

---

[*]Corresponding author E-mail: aditya4193@gmail.com

[8]. It is 100% transparent to visible light and has a properly large work function which stimulates the injection of holes onto the HOMO level of subsequent organic layer. It is preferred due to it having good optical transparency and seamless electrical conductivity. Having mentioning these key features, ITO also presents few major shortcomings, it is an expensive material (indium) because it is scarce , poor(large) refractive index, which results in power loss due to total internal reflection (TIR) not only at the ITO/glass but also at ITO/organic junctions, also it has bad mechanical sturdiness, which is not suitable for flexible display equipment [8].

Another potential candidate for anode material is graphene. A research undergone at Jilin University of china, along with Louisiana State University of Louisiana in 2012 shows that graphene depicted a better current efficiency and power efficiency than similar ITO-based devices working at a lesser current [7,8]. Graphene also has incredible physical, electrical and chemical properties, its atomic thinness leads to a high degree of flexibility and transparency, making its candidature strong for transparent electrodes [9]. Although, the graphene-based OLEDs have not shown any kind of improvement in efficiency over its counterpart ITO, they are till date very similar in performance to ITO based OLED devices.

Graphene too has its fair share of drawbacks one of which is a very low work function when compared to handful of conducting polymers, particularly poly (3, 4-ethylene dioxythiophene): poly (styrene sulfonate) also known as PEDOT: PSS. In multilayer OLED structure PEDOT: PSS is used as anode along with ITO (multi anode topography), it is generally used as hole injection layer (HIL) as it promotes charge transfer smoothly across different layers. PEDOT: PSS has gathered much attention for organic light emitting devices because it is cheaper than ITO as well as can be easily produced in mass [10]. Although both efficiency and stability of PEDOT: PSS have improved drastically over the years, still much improvement is required before their application in commercial display devices.

When treated with hydriodic acid (HI) in the presence of 0.01 wt.% GO hybrid film, PEDOT: PSS obtains a much higher conductivity than before [11,12]. The HI treatment helps in the expulsion of PSS chains out of the PEDOT: PSS thereby increasing the conductivity of the compound. The PEDOT which is now remaining together with the reduced GO sheets can provide more conductive routes for the carriers and increase the overall conductivity of the film [7,11,12]. It is worth mentioning that after being mixed with reduced GO sheets the open-air firmness of PEDOT is augmented drastically. The PEDOT: PSS/GO composite also provides a much better hole transportation (HTL) in organic photovoltaic (OPV) and organic light emitting diodes (OLED). However, one must note that the GO composite has an insulating property which can jeopardize the device performance hence a precise control of thickness of GO layer is much required.

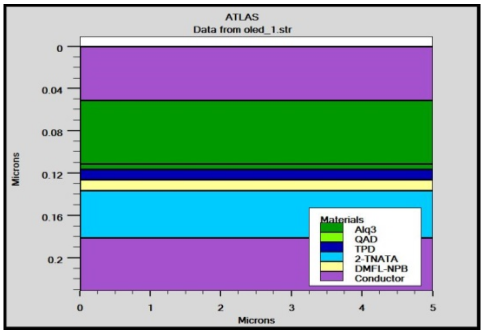

Fig. 1: The 5 layer structure of OLED.

Table 1: Specification of various layers used in proposed OLED structure.

SN	Layers used in OLED	Material	Material Thickness (nm)	Property
1	Hole injection layer	2-TNATA	45 nm	HOMO-LUMO (5.1-2.0 eV)
2	Hole transport layer	DMFL-NPB	10 nm	HOMO-LUMO (5.2-2.3)
		TPD	10 nm	HOMO-LUMO (5.4-2.4 eV)
3	Emissive layer	Alq$_3$: QAD	5 nm	HOMO-LUMO (5.5-3.5 eV)
4	Electron transport layer	Alq$_3$	60 nm	HOMO-LUMO (5.8-3.0 eV) [13]
5	Cathode	Al/LiF (fixed)	51 nm	Work function 3.34 eV
6	Anode	ITO	50 nm	Work function 4.70 eV
		Modified ITO	50 nm	Work function 4.83 eV [14]
		Graphene	50 nm	Work function 4.89 eV [15]
		Graphene oxide (GO)	50 nm	Work function 5.00 eV
		PEDOT: PSS	50nm	Work function 5.13 eV
		PEDOT: PSS/GO composite	50 nm	Work function 5.32 eV [10]

2 Device Details

In order to make the qualitative study effective and simple we are proposing a multilayer OLED device, with Anode/2-TNATA/DMFL-NPB/TPD/Alq3:QAD/Alq3/ Cathode, (see figure 1 for the proposed structure diagram and table 1 for the configurational properties of the mentioned elements) in the form of layers. It is to be noted that a multilayer device is preferred over single layer OLED due to its foreseen advantages like better hole transportation, hole blockage at cathode side and electron blockage at anode side. The proposed device is a multilayer device which is composed of five layers sandwiched in between electrodes(metals). The 4,4,4-Tris[2-naphthyl-(phenyl) amino]-tri-phenyl-amine (2-TNATA) is used as a HIL material due to its good adhesive nature to its neighboring layers. In the proposed structure we use two-hole transporting layers. One of them is TPD which is one of the most used HTL material. Another one is 9,9-Dimethyl-N,N-di(1-naphthyl)-N,N-diphenyl-9H-fluorene 2,7- diamine (DMFL-NPB) which is used to further enhance the movement of charge carriers. We have incorporated Alq3 as electron transport layer (EIL) because it has good electron mobility and has very good luminescence efficiency. The significance of EIL lies in the fact that it eases the flow of electrons from cathode into the recombination layer. EIL and HIL should be chosen wisely such that the HOMO of HIL should match the LUMO of EIL. A QAD doped Alq3 will further enhance the mobility of EIL while reducing the cut-off voltage. Indium tin oxide is employed as the electrode as a basis here. We then change the work function of the anode as per the selected candidate electrode materials for a thorough comparison and behavioral analysis of proposed OLED material. Also note that the cathode material is fixed to aluminum (Al) with a buffer layer of lithium fluoride (LiF) below it.

3 Results and Discussion

It has been foreshadowed in previous sections that changing the anode material greatly affects the device property and thus its performance. In order to analyse the performance we simulated the proposed device by using ALTAS SILVACO TCAD tool. This tool is used in order to better apprehend the internal behaviour of the device. In order to compare the various anode materials we use them consecutively in the proposed device. Every device structure has a different anode material and we have pitted them against each other to see which device has the best performance in terms of, luminescent power, current density (J), singlet exciton density, hole concentration and electron concentration.

First and foremost change which is seen in the device performance is its enhanced current density depicted in the J-V graph (figure 2(a)). It can be clearly seen that as we increase the work-function of anode from 4.7 eV to 5.32 eV the current density upsurges exponentially. The exponential increment with the voltage is in good agreement with the diode general characteristics. Moreover, for the analysis of conduction of designed OLED we have plotted the log-log J-V curve (figure 2(b)).

The graph can be divided into four different regimes as per the power law relation. The first is the ohmic regime for which the slop of the curve is “1”. In this the current is directly proportional to the voltage. The second regime is when the slope>1, known as space charge limited current (SCLC) with traps. In this the injected carriers consist of free and trapped ones. The third regime is known as trap filling limited current (TFLC) that indicates the trap states triggered by the contaminations. Traps are filled steadily with increasing voltage until the trap filled limit is attained. The fourth regime corresponds to the SCLC without traps. In this regime the current flows only because of the injected charge carriers. These all obtained regimes go well with the reported work on OLEDs [16]. Now as per the literatures the anode work-function should match with the HOMO of the HIL layer for high performance OLED device. From the JV curve we can clearly estimate that current density is minimum for 4.7 eV and maximum for 5.32 eV. Additionally, it can also be observed for work function greater than 5 eV there is no such significant increment in the current density. The reason can be attributed to the fact that PEDOT:PSS work function matches favourably to the HOMO energy level of HIL (2-TNATA) layer.

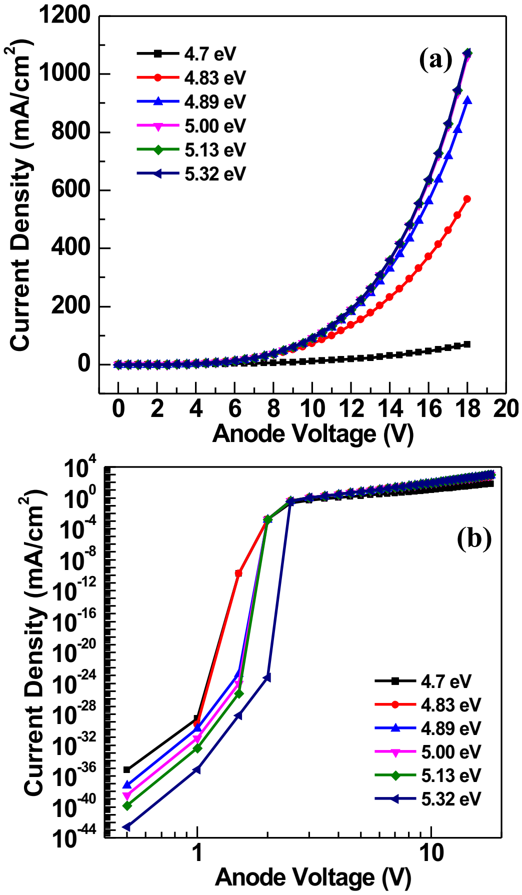

Fig. 2: Variation of current density with anode voltage in different anode material (a) Linear-linear Plot, and (b) Log-log Plot.

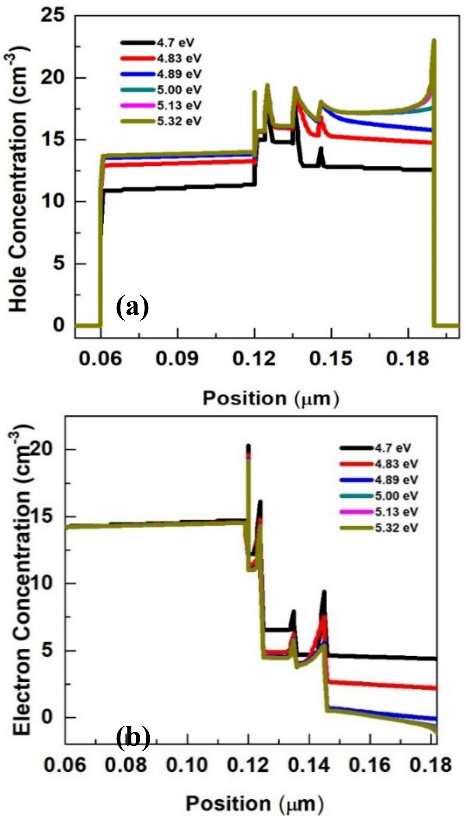

Fig. 3: (a) Variation of hole concentration with distance between electrodes, and (b) Variation of electron concentration with distance between electrodes.

Table 2: Recombination rates for different anode multilayer OLED.

S. No.	Work Function (eV)	Recombination rate ($cm^{-3}s^{-1}$)
1	4.70	4.83 x 10^{25}
2	4.83	3.16 x 10^{26}
3	4.89	4.72 x 10^{26}
4	5.00	5.43 x 10^{26}
5	5.13	5.48 x 10^{26}
6	5.32	5.49 x 10^{26}

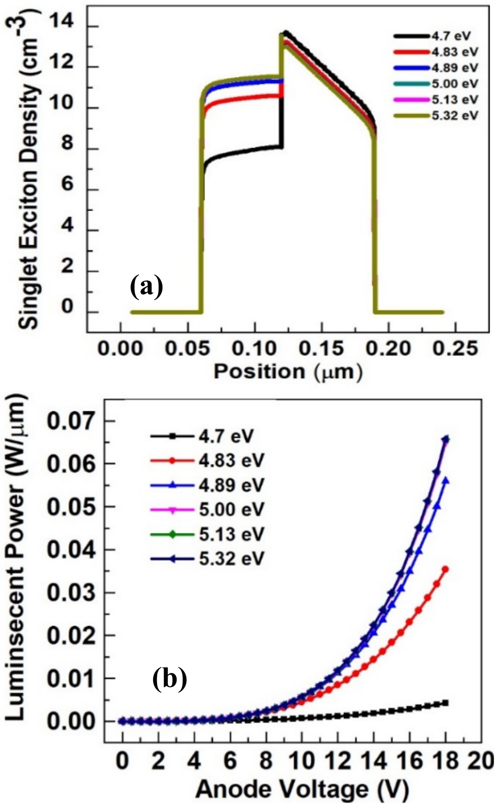

Fig.4: (a) Comparison of density of excitons in EML in different types of anode material used in OLED, and (b) Variation luminescent power with anode voltage in different anode material.

The concentration of holes plays an important role in the overall performance of the device, since we know from previous studies that the mobility of holes is greater than electron in OLEDs. In OLED the concentration of holes varies substantially when seen from cathode toward anode. The reason behind this is the concentration of hole is lesser near cathode (repels positive charges) and increases gradually as we move toward anode (attracts holes). As shown in figure 3(a), it can be clearly derived that the concentration of holes near hole transport layer (HTL) and emissive layer (EL) interface is more confined and it is maximum for the case of PEDOT: PSS/GO anode architecture, when compared to ITO and other anode candidate materials. Similarly, when we observe the electron concentration (in figure 3(b)), it gets reduced while moving from cathode to anode, but confines more near the emissive region.

Although in organic material the mobility of electrons is lesser than that of holes, the concentration of electrons plays a vital role for improving the recombination rate in EML (emissive layer). As the voltage is applied the hole and electron accumulate at the interface. The electron concentration in the active region is less because some holes get combined with the electron.

The recombination of hole and electron from same molecule recombine to form exciton. This exciton then generates photon. The singlet exciton density determines how much light will be emitted from EML (emissive layer). One exciton is capable of emitting single photon. If all the excitons are able to convert into photons, the device is said to have achieved 100% quantum efficient. As shown in figure 4(a) we can understand how significant the role of anode is (better work function) is in generating maximum amount of light. The recombination rate for diverse anode material is demonstrated in Table. 2.

Now as shown in figure 4(b), we can see a clear positive change in the luminescent power with respect to the same anode voltage across the device in case of each and every anode configuration. The performance of the device is the result of enhanced movement of charge carriers. More electron-hole pairs are reaching the emissive layer which is increasing the output light at higher voltages. Here the work function plays a vital role, we can observe from the graph that the ITO has lowest luminescent power, while the PEDOT: PSS/GO composite has the highest. ITO based OLED has lowest recombination rate and photon generation whereas PEDOT:PSS/ PEDOT:PSS composite Graphene-oxide based OLED shows the better recombination rate and the generation of photons among all. Thus high luminescent power for PEDOT:PSS/GO composite.

4 Conclusions

This paper highlights the key merits of using PEDOT: PSS/GO anode over other potential anode candidates. The higher work function of PEDOT: PSS/GO composite over ITO (almost 0.58 eV) clearly depicts an advantage. Though it is vital to properly manage the concentration of GO in PEDOT: PSS/GO composite anode. Too much GO will essentially make the anode an insulator, (recommended proportion of PEDOT: PSS and GO are 15:1). From all the results shown in this paper it can easily be concluded that PEDOT: PSS/GO composite anode can not only enhance the luminescent power with current density of the device but can also significantly improve the singlet exciton density in the device. The augmented performance enables us to use this anode material in OLEDs and various other opto-electronic devices.

Conflict of Interest

All authors declare that there is no conflict of interest regarding the publication of this paper.

References

[1] S. Kunić, and Z. Šego, “OLED technology and displays.” In Proc. ELMAR-2012, 31-35. 2012.

[2] A. Tiwari, B. Kumar, and A. K. Srivastava, “Computational study on CH3-functionalized Tetraphenyl-1, 4-phenylenediamine: A hole-transporting material for OLED devices,” Materials Today: Proceedings., **29**, 772-775, 2020.

[3] A. Tiwari, B. Kumar, and A. K. Srivastava, “Computational study on 8-quinolinolato-alkali, an electron transporting material for OLED devices,” In Proc. AIP Conference Proceedings., **2220**, 040019, 2020.

[4] A. Tiwari and B. Kumar, “Investigation of Different Layer OLED Structure based upon PEDOT: PSS/GO Composite Anode,” In Proc. 2020 International Conference on Electrical and Electronics Engineering (ICE3)., 583-587, 2020.

[5] S. Negi, P. Mittal, and B. Kumar, “Impact of different layers on performance of OLED,” Microsystem Technologies., **24**, 4981-4989, 2018.

[6] G. Tanghe, R. Thielemans, and N. Dedene, “Method and system for measuring and controlling an OLED display element for improved lifetime and light output,” U.S. Patent 7, Aug 28, 262 753, 2007.

[7] A. Uniyal, and P. Mittal, “Performance Comparison of Bilayer and Multilayer OLED,” Journal of Graphic Era University., **4**, 32-40, 2016.

[8] A. Srivastava, and B. Kumar, “Comparative study of graphene and its derivative materials as an electrode in OLEDs,” In Proc. AIP Conference Proceedings., **1952(1)**, 2018.

[9] A. Raj, M. Gupta, and D. Suman, “Simulation of Multilayer Energy Efficient OLEDs for Flexible Electronics Applications,” Procedia Computer Science., **152**, 301-308, 2019.

[10] Y. F. Liu, J. Feng, Y. F. Zhang, H. F. Cui, D. Yin, Y. G. Bi, J. F. Song, Q. D. Chen, and H. B. Sun, “Improved efficiency of indium-tin-oxide-free organic light-emitting devices using PEDOT: PSS/graphene oxide composite anode,” Organic Electronics., **26**, 81-85, 2015.

[11] X. Wu, L. Lian, S. Yang, and G. He, “Highly conductive PEDOT: PSS and graphene oxide hybrid film from a dipping treatment with hydroiodic acid for organic light emitting diodes,” Journal of Materials Chemistry C., **4**, 8528-8534, 2016.

[12] X. Wu, J. Liu, D. Wu, Y. Zhao, X. Shi, J. Wang, S. Huang, and G. He, “Highly conductive and uniform graphene oxide modified PEDOT: PSS electrodes for ITO-Free organic light emitting diodes,” Journal of Materials Chemistry C., **2**, 4044-4050, 2014.

[13] A. Srivastava, and B. Kumar, “Augmentation of OLEDs by Interpolating Electrode in the Hole Injection Layer,” In Proc. 2017 IEEE International WIE Conference on Electrical and Computer Engineering (WIECON-ECE)., 2017.

[14] J. T. Hu, K. L. Ye, Y. Huang, P. Wang, K. Xu, and X. H. Wang, “Effects of organic acids modified ITO anodes on luminescent properties and stability of OLED devices,” Optoelectronics Letters., **14**, 262-266, 2018.

[15] T. H. Han, Y. Lee, M. R. Choi, S. H. Woo, S. H. Bae, B. H. Hong, J. H. Ahn, and T. W. Lee, “Extremely efficient flexible organic light-emitting diodes with modified graphene anode,” Nature Photonics., **6**, 105-110, 2012.

[16] J. Steiger, S. Karg, and H. V. Seggern, “Electronic traps in OLED transport layers: Influence of doping and accelerated aging.” In Organic Light-Emitting Materials and Devices IV, International Society for Optics and Photonics., **4105**, 256-264, 2001.