International Science Journal (Peer-Reviewed)

ISSN: 2348 – 604X (p); 2348 – 6058 (e)

DAMA INTERNATIONAL

Review Article

GRAPHENE: A NOVEL MATERIAL AND ITS SMART APPLICATIONS IN ORGANIC LIGHT EMITTING DIODES

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ABSTRACT
A recently discovered graphene is going to be a most useful material for many applications in the areas of electronics, chemical, and biological sciences, etc. This novel allotrope of carbon has a two-dimensional sheet of mono-layer of carbon and unique band structure that makes it so important for utilizing in electronics especially in the Fabrication of Organic Light Emitting Diodes (OLEDs), Field Effect Transistors(FETs), photovoltaic cells, batteries, transistors, sensors and many other fascinating electronic devices. Beside this, graphene is going to become a promising material for its unique mechanical, optical, electrical properties. It is thinnest, strongest, stiffest, robust and the most conducting material in comparison to other known materials. This review traces the introduction of graphene in the field of organics electronics, particularly in the fabrication of organic light emitting diodes (OLED) and highlights the latest advancements and challenges including the applications of graphene as transparent electrodes, as hole injecting layers etc. Our analysis will be useful for those new researchers who are looking for the solution to make efficient, highly flexible, better light extraction OLED displays and energy-efficient, economical, solid state lighting devices.

KEYWORDS: Graphene, OLEDs, Transparent electrodes, Hole injecting layer

1.0 INTRODUCTION

Organic Light Emitting Diode (OLED) is an electroluminescent device based on organic materials that emit light under the application of external voltage. Its structure (100-500 nm thick) consists of a conductive layer, an emissive layer, sandwiched together between two electrodes and deposited on a substrate. There are two types of organic material are used in OLED: small organic molecules and long-chain organic polymers. Presently, OLED technology is growing very fast. It may be said, mainly on the basis of two facts, one by seeing the time span (1990-2000) where OLEDs has achieved the performance similar to the unfiltered incandescent lamp - almost one third time less than the conventional LEDs and second by increasing commercial activities its area. OLED displays and solid state lighting are being attracted by the consumers due to its light weight, brightness, colour quality, wide angle, flexibility, and transparency.

Research and development work, on OLED, was started during 1950-60 when some organic molecules showed high fluorescence quantum efficiency. The first study of electroluminescence (EL), was done by Bernanose et al. (1950), using dispersed polymer film. The curiosity of researchers increased when Pope et al. (1963) performed the study of electroluminescence with anthracene crystal. The expected breakthrough came in year the 1987, when Tang and Van Slyky (1987) demonstrated a highly efficient multilayer OLED with a quantum efficiency of 1% and luminescence 1 lm/w by using Alq_3 (small molecule) as the emitter material and transparent Indium Tin Oxide (ITO) as a cathode. This device was fabricated using chemical vapor deposition (CVD) technique. Results from this experiments were enough to start the commercial activities. In the year 1990, Burroughes et al. (1990) used first polymer (long chain molecules) which enriched the technology by providing the idea of using solution processed thin layer of a

ISSN: 2348 – 604X (p); 2348 – 6058 (e)

conjugated polymer as an emitter material. With this, concepts like inkjet printing and roll to roll processing of OLED emerged. Baldo et al. (1998) used phosphorescent dyes and with this attempt internal quantum efficiency of organic light emitting diode could reach up to 100%. Further in year the 2006, Sun et al. (2006) used a blue fluorescent with green and red phosphor dopants. OLED efficiency and lifetime further get enhanced by more than 100 lm/w and more than 100000 hours respectively by introducing a concept of electrical doping of transport layers. With these advancements, application of OLEDs was started in small-screen devices such as mobile phones, digital cameras, and PDAs. Kodak introduced a digital camera with an OLED display in the year 2003, Sony Corporation started the mass production of OLED screens in 2004, and Samsung Electronics declared that it had developed a prototype 40-inch, OLED-based, ultra-slim TV in the year 2005.

In search of material suited for efficient, highly flexible, better light extraction OLEDs displays and energy-efficient, solid-state lighting devices, researchers focused their mind towards carbon nano-material for transparent electrodes, hole transport layer (HTL), electron transport layer (ETL) of OLEDs. Carbon nanotubes (CNTs) - a hollow cylinder of graphite sheet (three- dimensional shape) attracted more because of its high aspect ratio (~1000), unique electrical, optical and mechanical properties. These CNTs are being used in the form of single-wall nanotubes (SWNT) and multi-wall nanotubes (MWNT) and have been found to act as a hole blocking material, efficient hole injectors, and transparent electrodes. Light extraction from top and bottom was made possible due to its large surface area, three-dimensional hole injection, flexibility, room temperature processing and good acid resistance. Several studies are being carried out on CNTs based OLEDs even today to improve the performance further. Perhaps, these results with CNTs would have been one of the motivational factors for researchers for introducing graphene in OLEDs.

2.0 Graphene and its specialty:

Graphene became the point of attraction for the researchers when Andre Giem in 2004 discovered this novel nanostructure material by isolating single layer graphite on an insulating layer of (SiO_2)[4]. Graphene is allotropes of carbon and has a layered structure (Fig.-1). The distance between two layers is 0.335 nm . In each layer or plane, carbon atoms are arranged in a hexagonal shape with the distance of 0.142 nm and have a covalent bond with sp^2 hybridization that makes it very strong. These layers may be separated because each plane are bound together with weak Vander Waals forces. This single hexagonally, two-dimensional layered of carbon is called graphene. Other allotropes of graphite are fullerenes (zero-dimensional), Quantum dots (zero-dimensional) and carbon nanotubes (one-dimensional) etc. (Fig.-2). Graphene is highly transparent and absorbs only 2.3 % of light falling on it which makes it idle to make transparent electrodes. Other important characteristic includes, its high intrinsic carrier mobility (200000 cm^2v^{-1} s^{-1}), the high optical transmittance of 97.7%, high Youngs modulus of 1.0 TPa. Graphene has an ability to sustain extremely high electric current density as compared to copper. This great conductor does not have a band gap, means it cannot be switch off. It has been proved 200 times stronger than steel and stretchable up to 20% of its original length without getting a break. These properties are therefore being utilized in making the flexible, unbreakable displays. Graphene is also behave like a excellent heat conductor.

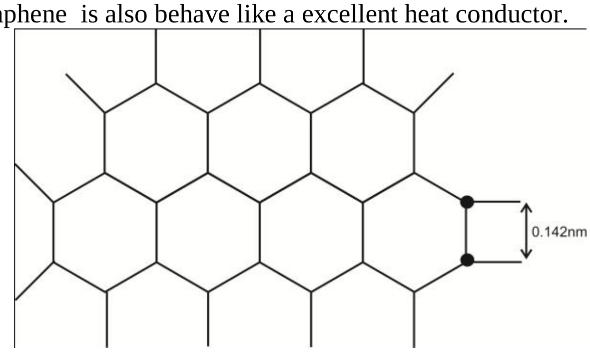

Fig : 1Two-dimensional hexagonal sheet of graphene

ISSN: 2348 – 604X (p); 2348 – 6058 (e)

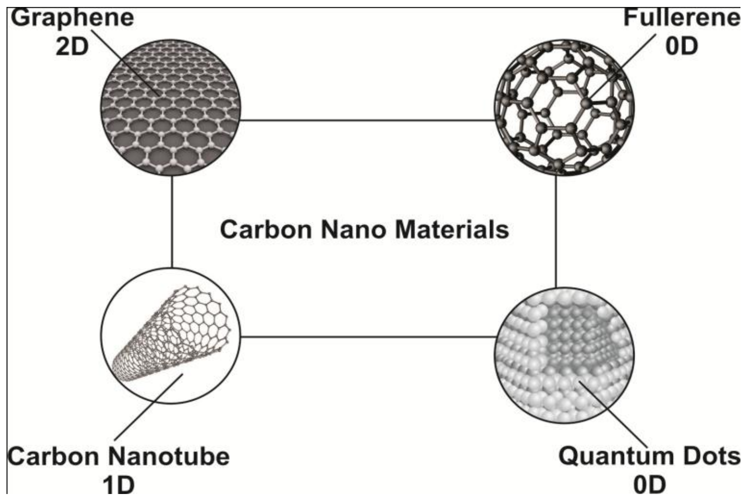

Fig : 2 Carbon nano-materials

3.0 Application of Graphene in OLEDs

Graphene is being employed in Organic Light Emitting Diode as a transparent electrode or as a layer for injecting hole directly into the emissive layer, presently.

3.1 Graphene as Transparent Conducting Electrodes

Idea of introducing, graphene electrodes,(fig.-3) came into the mind of the researchers by knowing its remarkable properties such as its high electronic mobility, optical transparency, and flexibility.

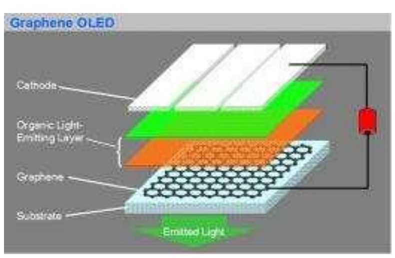

Fig.-3 : Graphene as a electrode in OLED structure.
Source: http://phys.org/news/2010-03-nanometer-graphene-oleds.html

Although, *Indium Tin Oxide* (ITO)- an conventional transparent electrode, still most widely used for such applications because of its good transmittance in the visible (>80 %) and near IR regions and for its low electrical resistivity(~20 Ω/□), etc.. Beside these advantages, it also have certain limitations such as its brittle nature, density, high processing temperature, imperfect work function, poor transparency in blue region, susceptibility for corrosion, limited availability of Indium in nature and its high price . Hence, the scientific community is looking graphene as a most potential transparent electrode for the next generation of devices. In the year 2009, Wu *et al.(*2009) fabricated solution processed thin film of graphene oxide thin film using Hammers method and found a sheet resistance of about 800 ohm/sq and 82.0% transmission at 550 nm. These results were found below the expecting values and reason was

International Science Journal (Peer-Reviewed)
ISSN: 2348 – 604X (p); 2348 – 6058 (e)

given that during the fabrication process some defects like lattice defects and multiple gain boundaries etc were formed. Although the solution based graphene started its operation at 4.5V and showed a luminance of 300 cd/m^2 at 11.7 V. (ITO starts operation at 3.8 V and show luminance 300cd/m^2 at 9.9 V). These results were improved further by Bae *et al*. (2010) using a single layer of graphene and demonstrated a sheet resistance of 125ohm/sq and 97.4% transmission at 550 nm. These results were found more promising then ITO and other transparent conducting materials. The same year in the year 2010, Sun *et al.* (2010) performed the study with multilayered graphene using CVD and found that transmittance of graphene electrodes decreases with increasing of layers. Device performance was not up to mark as ITO. Rough surface and unaligned work function were the main reasons for low efficiency and high leakage of current. Hawg *et al. (*2012) gave the solution for this problem using weak plasma treatment and improved the performance of multilayered (without polymer interface layer) graphene. OLEDs with small molecules fabricated by vacuum deposition method that is quite easy and effective. On the other hand, polymer-based OLEDs are fabricated by generally solution processes such as spin coating, ink-jet printing, screen printing, dip coating and dip coating but top electrode has still been fabricated by vacuum deposition. Recently Jung-Hung Chang *et al*. (2015) fabricated top electrode cathode by solution-processed methods with using n-doped graphene as top cathode and results are shown in (Fig.-4).

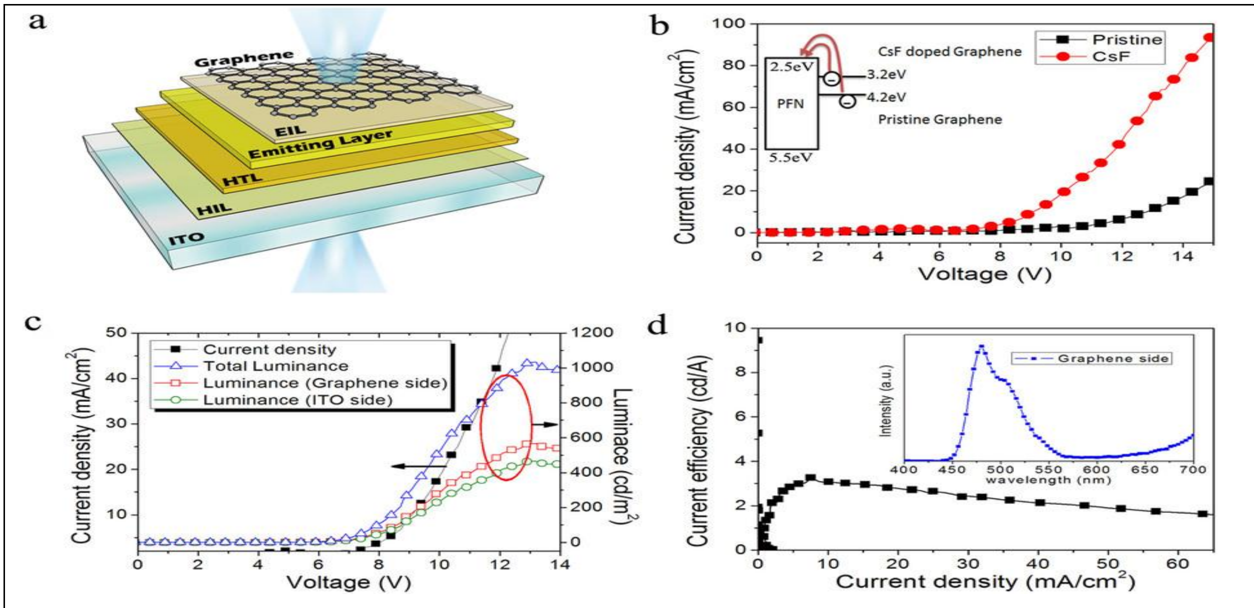

Fig: 4 (a) The device structure: ITO/HAT-CN/VB-FNPD/PVK:Firpic/PFN:CsF/multilayer graphene, (b) The current density versus voltage (J-V) curves of devices with 4-layer pristine graphene and 4-layer CsF-doped graphene as a cathode. The inset shows the schematic illustration of electron injection from pristine graphene or CsF-doped graphene. (c) The current density-voltage-luminance (J-V-L) curve of the transparent polymer OLEDs, (d) Luminance efficiency verse current density characteristics. The inset shows electroluminescence spectra of the blue-light transparent polymer OLEDs from graphene cathode side.
Source: *Scientific Reports* 5, Article No: 9693, 2015

Due to thinnest in nature, graphene electrodes found suited for better light extraction (no reflection and no trapping) due to its. Generally in ITO light reflected from the top and bottom interface and much amount of light trapped. This is why more light coupled out from graphene- based OLEDs.

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ISSN: 2348 – 604X (p); 2348 – 6058 (e)

In solid state lighting in the year 2013, power efficiency 80 lm/W at a high brightness of 3000cd/m^2 was demonstrated using graphene-based white organic light emitting diode (WOLED's) by Li et al., in the year 2013. Results were quite significant because the demonstrated efficiency was more than the incandescent bulb (average 12-17 lm/W) and comparable to both fluorescent lamp (> 80 lm/W) and inorganic white LEDs (> 100 lm/W).

Critical challenges for graphene-based OLEDs is not only to the improvement of large area but also to improve efficient charge injection and graphene doping. Several dopants were tried further but due to unstable in nature, most of them were not suited for the application in OLEDs. In the year 2014, Jens Meyer et al. (2014), used an intermediate layer of molybdenum trioxide (MoO_3) between a graphene electrode and HTL and found that few nanometer of MoO_3, a p-type dopant is sufficient to improve the sheet resistance up to three times. OLEDs with structure, Graphene/ MoO_3/HTL/EL/EBL/ETL/Al, showed small barrier for hole- injection but other electrical and optical performance found similar to the ITO- based devices (Fig.-5).

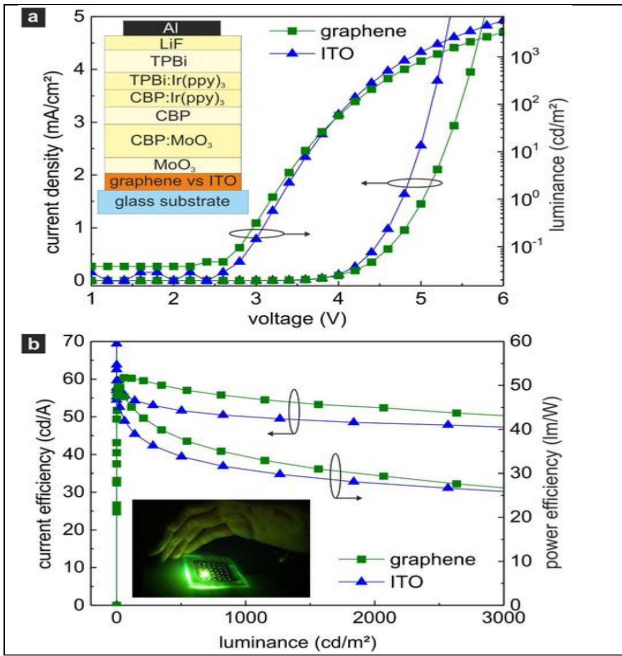

Fig : 5 (a) Current efficiency Vs voltage characteristic of OLED with ITO and Graphene electrode respectively

(b) Current efficiency (cd/A) and power efficiency (lm/W) Vs luminance. Photograph in inset shows the OLED at high brightness level.

Source: *Science Reports* 4, Article Number:5380 (2014)

Table 1. Comparative study of different transparent electrode materials for OLED's

Materials	Conductivity/ Transparency at 350nm	Demonstrated OLED Performance	Light Extraction	Reliability	Mechanical Flexibility	Challenges to replace ITO
ITO	10 Ω/sq at 90%	>100 lm/w	Light trapping ITO	Good	Poor	-
CNTs	500 Ω/sq at 85%	10cd/A at 1000 cd/m^2	Medium	Excellent	Flexible/ Stretchable	Conductivity Roughness cost
Metal Nanowire	30 Ω/sq at 93%	54 lm/W similar to ITO control device	High Angle Uniformity	Good	Flexible	Stability cost
Polymer	39 Ω/sq at 80%	12% EQE similar to ITO	High	Medium	Flexible	Conductivity stability
Graphene	125Ωsq 97% 30Ωsq 99%	103 lm/W for Green 80 lm/W for white	High (No reflection/ No Trapping)	Excellent	Most Flexible/ Stretchable	Conductivity cost

Source: Tze-Bin Song, Ning Li. *Electronics* 2014, 3, 190-204.

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3.2 Graphene in Hole Injection Layer

The performance of OLEDs mainly depends upon charge injection from electrodes. The various hole injecting layers and technique were introduced to improve the hole injection. Shengwei Shi et al. (2013) demonstrated a high luminance of over 53000cd/m^2 at 10 V and record luminance of 40,785 cd/m^2 at only 8.8 V which was 30 times more than the reference device (fig.-7). This device was fabricated on ITO with N, N-bis (3-methyphenyl)-N,N-diphenyl-1,1-biphenyl-4, 4-diamine (TPD) as the hole transport layer and Tris (8-hydroxyquinolinato) aluminum(Alq_3) as the electron transport and emissive layer and solution- process GO was used as a hole injecting layer (fig.-6).

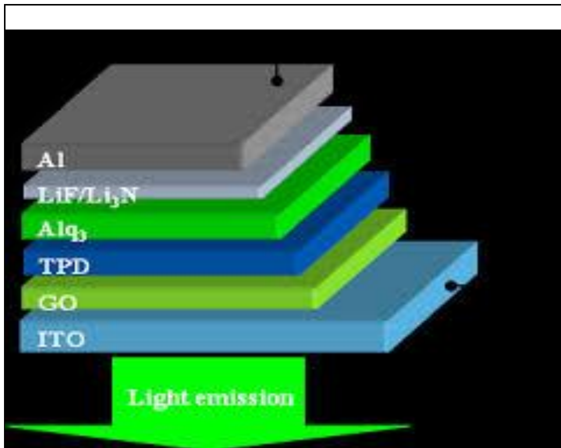

Figure 6. Scheme of OLED device structure
Source: *J. Mater. Chem.* C, 2013,1, 1708-1712

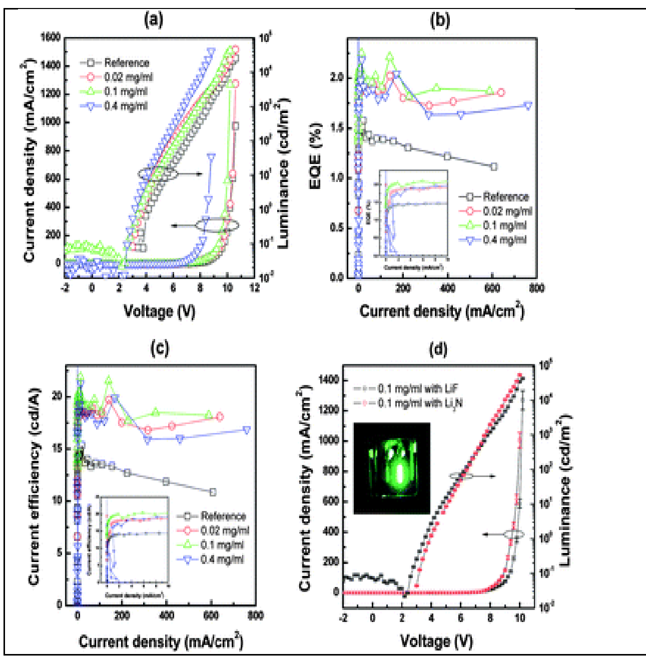

Fig:7
Device performance for OLEDs with pure and modified ITO by GO in different concentrations. (a) Current density-Voltage-Luminance, (b) EQE-Current density, (c) Current efficiency-Current density, and (d) The comparison of Current density-Voltage-Luminance characteristics for OLEDs on GO/ITO anode with different electron injection layers, inserted is a real-time picture during device operation.
Source: *J. Mater. Chem*. C, 2013,1, 1708-1712

In polymer-based OLEDs, PEDOT:PSS [Poly(3,4-ethylenedioxythiophene): poly (styrene sulfonate)] is widely used as hole transport layer. Graphene has a potential to replace the PEDOT:PSS because it never reacts with any material. On the other hand PEDOT: PSS degrade the electrodes due to its acidic nature particularly in the presence of moisture.

4.0 Problem and finding associated with the Graphene in OLEDs

Graphene offers a remarkably high electronic mobility. Even though the concentration of such carriers is low which leads to its unimpressive performance as an electrode (high carrier mobility observed only 3000cm^2/Vs, sheet resistance >1kΩ but the theoretically it should be > 150,000 cm^2/Vs). This problem may be rectified by doping the graphene layer with excess carriers. Actually by doing so, free carrier density and work function for hole injection (work function 4.5eV is too low as an anode for hole injection) will increase. Here, it is necessary that while doping high optical transparency of graphene must be preserved. Beside the above by improving the coupling technique (with lens) external quantum efficiency may be improved. So optimization is required for better light extraction.

5.0 Conclusion and perspectives

Research works are being carried out at a different level on graphene-based OLED, throughout the word. In the initial phase, results shown by various research groups are magnificent . Till now graphene has been found to act as an electrode, an efficient hole injecting layer and have also been tried in to make a top electrode. The best results, regarding the application of graphene in OLED are found when it was used as a transparent electrode. These results show distinct advantages over popular ITO- based electrode. It would have been too early to say that graphene is the most efficient material with regards to transparent electrode but its properties of most flexible, better light extraction capability, stability, compatible with other organic material and cost reduction potentiality using large area roll to roll processes make it extraordinary material to replace the conventional ITO.

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