Carbon Dots: The future of display and lighting applications

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ABSTRACT

We demonstrate the ability to manufacture Carbon Dots Light Emitting Diode (CD-LED) devices using three process layers of all solutions with either conventional or inverted structures for red, green, blue and white emitting devices. This represents the cheapest method of manufacturing the display and lighting panels.

1 Introduction

Light Emitting Diodes (LEDs), now extensively used in consumer electronics, are considered as part of a key transformation in the field of displays and lighting. These have high current to light efficiency, improved colour contrast and are able to make larger panels than conventional liquid crystal displays.

Since 1987, organic light-emitting diodes (OLEDs) have attracted considerable interest owing to their promising applications in flat-panel displays by replacing CRTs or LCDs. However, OLEDs' production cost are expensive, as it needs a high vacuum for thermal evaporation (VTE) fabrication techniques, making the material cost high due to the additional high wastage cost and the significant power usage for operating rotary pumps and cryopumps. The new demanding display specification ITU-R-BT-2020 (REC 2020) is for high definition TVs. The usual fluorescent and phosphorescent emitters only achieve roughly 90% of NTSC standards (National Television System Committee) developed in 1941, for black-and-white television and then in 1953 a second version of NTSC standard was adopted, which allowed for colour television broadcasting[1]. It is not possible to satisfy this by OLEDs, so an alternative emitter was sought, leading to the emitter replacement with colloidal quantum dots (QDs)

Colloidal quantum dots (QDs), made of inorganic materials with core and shell structure can be solution processed. QDs have found commercial applications as optically excited colour enhancers. However, due to the cost of synthesizing QDs and their toxicity, we have sought an alternative material, specifically Carbon Dots (CDs) are being investigated for this purpose.

Carbon dots (CDs) have fascinating strong physicochemical nature, including size less than 10 nm, rich functional groups, strong Photo Luminescence (PL) and Electro Luminescence (EL), biocompatibility and non-toxicity (Shou Li et al, 2021). CDs have been acclaimed as another remarkable carbon-based nanomaterial like fullerene, nanotubes and graphene. The evolution of CDs can be divided in three stages; first is the discover stage, from 2004 to 2006, second is between 2007 and 2011, developing stage, when CDs were synthesised, and the final application stage from 2011 to now, where research has expanded into discovering possible applications.

CDs were first reported and shown by Xu et al as fluorescent nanomaterial[2], but they did not attract other researcher's interest, until Sun et al formally defined them as Carbon Dots in 2006[3]. In the developing stage, CDs were generated from diverse carbon containing materials, e.g. citric acid to graphite, as precursors which give different optical and electrical properties by varying experimental parameters [4-15].

Figure 1 shows the side-by-side Red/ Green/ Blue CDs pixel array Device structure for Displays and Signage purpose and the White Carbon Dots Light Emitting Device tandem three stack structure for Lighting panels.

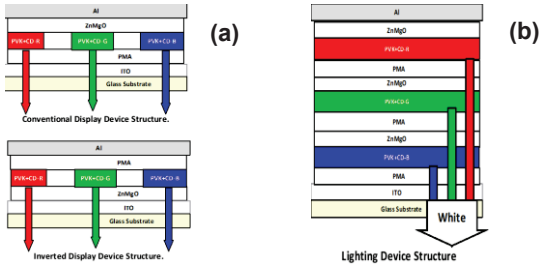


Figure 1. (a) Conventional and Inverted device structure for Display. (b) Conventional Tandem device structure for Lighting panels.

Conventionally, displays using Organic Light emitting Diodes (OLEDs) can be configured as either Passive Matrix (PMOLED) or Active Matrix (AMOLED). PMOLED displays require each row to be controlled sequentially resulting in restricted overall brightness and limited array size. AMOLED displays incorporate TFTs and capacitors to mitigate the restricted brightness of sequential row control, this in turn enables arrays of arbitrary size to be designed with realistic parameters. Accordingly, AMOLED matrices represent the overwhelming majority of the market for OLED display technology.

2 Experimental

In this section we will describe the details about synthesis of carbon dots, ink formulations and device fabrication.

2.1 Synthesis of Multi-colour CDs

CDs were synthesized by standard published methods [4,8,11]. The figure 2 shows the blue, green and yellow CD solutions made at Brunel University London under room light and UV light.

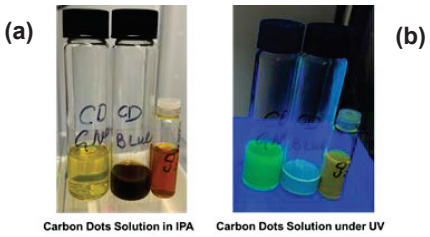


Figure 2. Blue, green and yellow CD solutions made at Brunel, (a) under day light and (b) under 365nm UV,

2.2 Formulation of inks

30mg of Phosphomolybdic acid (PMA), (Luminescence Technology Corp., Taiwan), was dissolved in 10ml of isopropanol and used for the Hole Injecting layer (HIL).

50mg of Polyvinyl carbazole (PVK), (Luminescence Technology Corp., Taiwan), and 50mg of CDs, synthesised at Brunel University, were dissolved in 10ml of 1,4-Dioxane and used as the emissive layer (EL).

Zn $(OAc)_2.2H_2O$ (180mg, 0.1M) and $CH_3COO)_2Mg \cdot 4H_2O$ (40mg, 0.1M) (Sigma Aldrich), Monoethanolamine (60 mg, 0.1M) and anhydrous ethanol (10ml) were added to a round bottom flask and refluxed at 80°C for 1h to form a clear homogeneous solution of ZnMgO which was used as an electron transporting layer (ETL).

2.3 Device fabrication

The patterned ITO substrates were successively ultrasonically washed with acetone, isopropyl alcohol and deionised water, each for five minutes. The ITO substrates were then dried in a vacuum oven for twenty minutes at 120°C then treated with UV-ozone for ten minutes and finally moved to the plasma treatment chamber and exposed to an oxygen plasma for twenty-five seconds before use.

For conventional devices, the PMA ink used as HIL was spin coated onto ITO glass at 3000 rpm, followed by baking at 200°C for 15 mins. Thereafter, the emissive layer of PVK+CDs was spin-coated at 2000 rpm, and annealed at 120°C for 30 mins under vacuum. Then a sol-gel ZnMgO solution was deposited on the CDs layer at 3000 rpm and annealed at 120°C in air. Finally, a 100nm aluminium (Al) electrode was thermally evaporated under high vacuum in the Solciet.

For inverted devices, the ZnMgO sol-gel solution (ETL) was spin coated onto ITO glass at 3000 rpm, followed by baking at 200°C for 15 mins. Thereafter, the emissive layer of PVK+CDs was spin-coated at 2000 rpm, and annealed at 120°C for 30 mins under vacuum. Next, a PMA ink was deposited on the CDs layer at 3000 rpm, and annealed at 120°C in air. Finally, a 100nm aluminium (Al) electrode was thermally evaporated under high vacuum in the Solciet.

All other layers including TCTA, α-NPB and TPBi were evaporated (1 As^{-1}) and HATCN (evaporated at 0.5 As^{-1}) in an ULVAC OLED plant (Solciet, purchased from ULVAC, Japan) onto a patterned ITO/glass (100 mm x 100 mm) under a vacuum of 10^{-5} Torr. Deposition of the cathode also took place under vacuum ahead of device encapsulation in a nitrogen-filled glove box.

We fabricated devices based on semi or fully solution processed either conventional or inverted structures.

Type A: ITO/ZnMgO (30 nm)/PVK+CD (25 nm) /TCTA (10 nm)/α-NPB (20 nm)/ HATCN (10 nm)/Al, Inverted and semi solution processed device structure.

Type B: ITO/PMA (10 nm)/ PVK+CD (25 nm) /TPBi (30 nm)/ LiF(0.5 nm)/Al, Conventional and semi solution processed device structure.

Type C: ITO/ZnMgO (30 nm)/PVK+CD (25 nm) /PMA (10 nm)/ Al, Inverted and fully solution processed device structure.

Type D: ITO/PMA (10 nm)/ PVK+CD (25 nm) / ZnMgO (30 nm)//Al, Conventional and fully solution processed device structure.

3 Results and Discussions

In this section we will report the thin film characterisation and the device performances.

3.1 Thin Film characterisation

3.1.1 Figure 3, shows the emission spectra of the blue, green and yellow CD thin films after spin coating at 2000 rpm and annealing at 120°C under vacuuming.

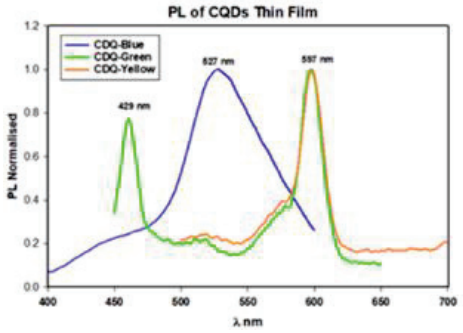


Figure 3. Carbon Dots (CDs)'s PL spectra

3.1.2 Figure 4 shows ZnMgO's absorbance spectra, AFM and SEM images. Sol-Gel ZnMgO and thin film made @ 3000 RPM, Film thickness: 32 ± 0.5 nm and Roughness, R_a= 1.3± 0.6 nm

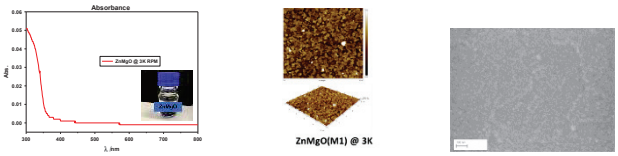


Figure 4. ZnMgO's absorbance spectra, AFM and SEM images.

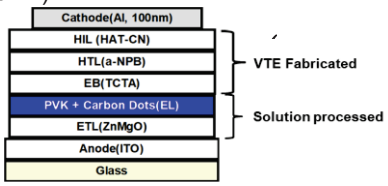
3.1.3 Figure 5 shows PMA's absorbance spectra, AFM and SEM images. Dispersion of PMA and thin film made @ 3000 RPM, Film thickness: 11 ± 0.5 nm and Roughness, RA= 1.9± 0.5 nm

Figure 5. PMA's absorbance spectra, AFM and SEM images

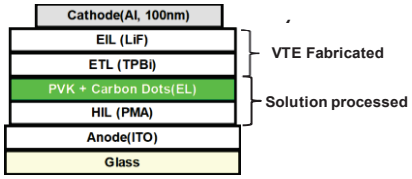
3.1 Device performances

Figure 6, shows device structural diagrams and figure 7, shows devices' performance plots and EL spectra. Finally, table 1, shows the summarised device performances. Unfortunately, device type D, which is fully solution processed and conventional OLED structure had very weak emission and could not be measured and we are investigating binder properties to understand this issue.

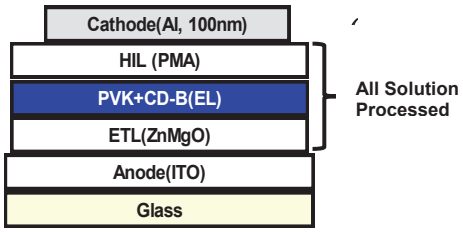
a) **Type A:** ITO/ZnMgO (30nm)/PVK+CD-B (25 nm) /TCTA (10 nm)/α-NPB (20 nm)/HATCN (10nm)/ Al



b) **Type B:** ITO/PMA (10 nm)/ PVK+CD-B (25 nm) /TPBi (30 nm)/ LiF(0.5 nm)/Al



c) **Type C:** ITO/ZnMgO (30 nm)/PVK+CD-B (25 nm) /PMA (10 nm)/ Al



d) **Type D:** ITO/PMA (10 nm)/ PVK+CD-B (25 nm) / ZnMgO (30 nm)/Al

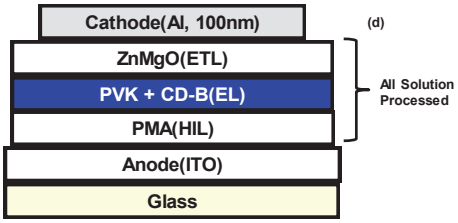


Figure. 6 The structure of **Type A –** partially solution processed Inverted structure **(a), Type B -** partially solution processed conventional structure **(b), Type C -** fully solution processed inverted structure **(c) and Type D -** fully solution processed conventional structure (d).

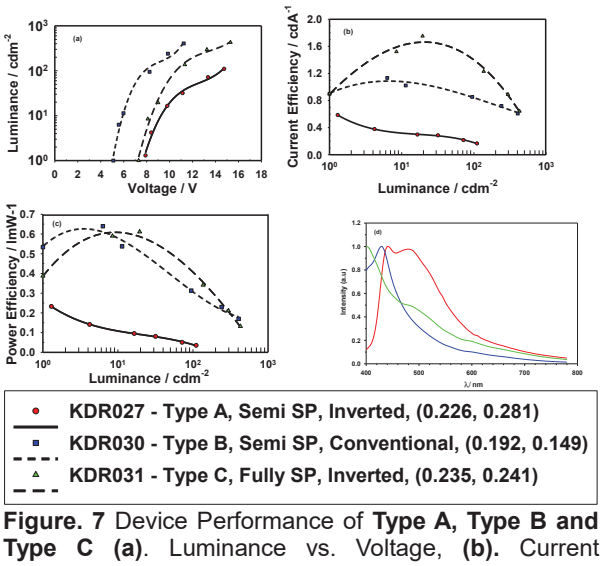


Figure. 7 Device Performance of **Type A, Type B and Type C (a).** Luminance vs. Voltage, **(b).** Current Efficiency vs. Luminance., **(c).** Power Efficiency vs. Luminance., **(d).** Electroluminescence spectra

Table 1. Device Performance for CD-LED devices **of Type A, Type B** and **Type C.**

ID	Type	V_T (V)	@ 100 cd/m²				Maximum		
			V_D (V)	C/E (cd/A)	P/E (lm/W)	CIE (x, y)	L (cd/m²)	C/E (cd/A)	P/E (lm/W)
KDR027	A	7.9	14.2	0.2	0.1	(0.226, 0.281)	150.3	0.6	0.2
KDR030	B	5.3	8.6	0.9	0.3	(0.192, 0.149)	405.0	1.2	0.6
KDR031	C	7.3	10.4	1.4	0.4	(0.235, 0.241)	428.5	1.8	0.6

4 Conclusions

We are able to make Carbon Dots Light Emitting Diode (CD-LED) devices containing three solution processed layers. We achieved maximum current efficiency of 1.1 cdA^{-1} and power efficiency as 0.6 lmW^{-1} for Blue Carbon Dots doped with PVK light emitting diodes where the device was made with three spin coated layers. We are working on optimising the device composition and processing parameters.

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