Solution-Processed Indium-Gallium-Nitride (InGaN) Blue Light-Emitting Diodes (LEDs)

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ABSTRACT

A soluble form of InGaN was synthesized, producing a solution that exhibited blue photo-luminescence (PL) upon UV light exposure. This solution was used in the fabrication of a blue light-emitting diode (LED) that was produced using solution processable methods, a world first for an InGaN-base LED. The PL properties of the solution and the electro-luminescence (EL) and device characteristics of the LED are presented.

1 INTRODUCTION

In recent years, the efficiencies and lifetimes of red and green organic light-emitting diodes (OLEDs) based on phosphorescent, thermally activated delayed fluorescent (TADF) and polymer materials have increased steadily. This has allowed for a broad expansion in the application of these materials, and they have become ubiquitous in the smartphone and flat panel display market. In contrast, a comparable improvement in the performance of blue-emitting OLEDs has not been observed, and engineering solutions have been developed to overcome their limitations for particular applications. However, for the blue emitters to reach performance parity with their red and green analogues, a new class of materials is required [1-3].

InGaN has already been widely utilised in blue-emitting LEDs. However, the process required to deposit InGaN require high temperatures, which obviates its deposition on glass or heat-sensitive plastic substrates. Also, the need to deposit via capital-intensive vacuum-based deposition techniques can limit the size of the substrates. This is in contrast to solution-based techniques that allow for the affordable and large-scale deposition of optoelectronic devices, as demonstrated by the solution-deposition of the light-emitting layers in OLEDs.

There is considerable academic interest in preparing a material that has the robust physical and optoelectronic properties of InGaN, but is also compatible with facile, solution-based deposition techniques. To this end, cadmium-selenide/zinc-sulfide (CdSe/ZnS) nanoparticles have been incorporated into blue-emitting LEDs, but these materials are highly instable against oxygen and moisture, severely limiting their application [4]. Furthermore, the presence of highly toxic cadmium creates concerns regarding the treatment of waste during the synthesis of the material and the disposal of consumer electronics at the end of their lifetime.

In this talk, we present the fabrication of such a solution from an InGaN powder precursor and demonstrate its application in the solution-based fabrication of a blue LED, and report the performance of such a device. We believe this presents a significant advancement in the field of inorganic LEDs.

2 EXPERIMENTS

2.1 Device fabrication

The InGaN-based solution was made from an InGaN powder using a proprietary method to be disclosed at a later date.

Figure 1 shows the device structure of the blue-emitting LEDs fabricated using a solution-processable InGaN-based emissive layer.

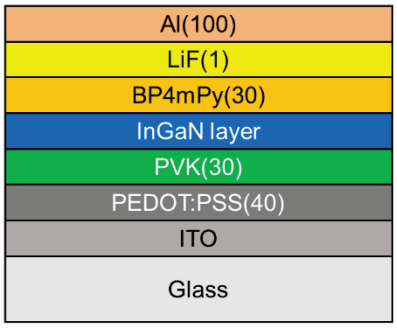


Fig. 1 Device structure of solution processed InGaN LED

Indium-Tin-Oxide (ITO, 110 nm thick, 10 Ω/□ sheet resistance) coated glass substrates (25 x 25 mm^2) were sequentially cleaned in an ultrasonic bath with de-ionized water and IPA, and treated by an UV-ozone exposure system (Novascan PSD Pro) prior to use.

A hole injection layer of 40 nm-thick poly(3, 4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS, Clevios 4083) layer was spin-coated onto the ITO at 3,000 rpm for 30 sec and baked at 220 °C for 15 min on a hot plate in air. A hole transport layer of 30 nm-thick poly(9-vinylcarbazole) (PVK, Lumtec LT-N4077) layer was also spin-coated using a PVK (20mg) /chlorobenzene (2 mL) solution at 4,000 rpm for 30 sec on the PEDOT:PSS layer and baked at 170 °C for 15 min on a hot plate in a nitrogen atmosphere.

The emissive layer was formed by spin coating a

proprietary InGaN-based solution at 1,500 rpm for 30 sec on the PVK layer and baking at 150 °C for 15 min in a nitrogen atmosphere.

The electron transport layer of 30 nm-thick 3,3',5,5'-tetra[(M-pyridyl)-phen-3-yl]biphenyl (BP4mPy, Lumtec LT-N862) was deposited in high vacuum of around $1X10^{-5}$ Pa on the emissive layer. The hole electron injection and cathode layers of 1 nm-thick lithium fluoride (LiF) and 100 nm-thick aluminum (Al) layers, respectively, were also deposited by high vacuum.

Finally, the devices were completed by the application of an encapsulation glass and desiccant using a UV curable epoxy resin. Each glass substrate had 6 pixels with 2 x 5 mm luminescence area for each pixel.

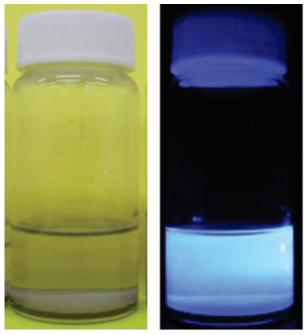
2.2 Device analysis

Current - Voltage (I-V) and Luminescence - Voltage (L-V) characteristics were analyzed by a Keithley 2400 source measure unit (SMU) and Topcon BM-7 luminescence colorimeter, respectively. The electro luminescence (EL) spectra was obtained using an ASEQ Instruments LR1 spectrometer.

3 RESULTS

3.1 PL from InGaN-based solution

Our synthetic method results in a colourless transparent solution containing InGaN. Figure 2 shows the solution used in the fabrication of blue LEDs under white light and UV light, with blue emission evident under the latter.



(a) (b)

Fig. 2 Photographs of InGaN/IPA solution under (a) white light and (b) UV light

As shown in Figure 3, we obtained a PL emission spectrum of the InGaN-based solution with excitation at 250 nm. In comparison with the InGaN source powder, the intensity of the emission is greatly enhanced. The spectrum has an emission peak at 427 nm with a wide full width at half-maximum (FWHM) of 102 nm.

3.2 EL from InGaN-based LED

Figure 4 shows the V-I-L characteristics and emitting top view (inset) of an InGaN-based LED. Figure 5 also shows the EL spectrum of the InGaN-based LED at a voltage of 7 V. It has an emission peak at 460 nm with a FWHM of 158 nm. The results at current densities of 1 and 10 mA/cm^2 are shown in Table 1.

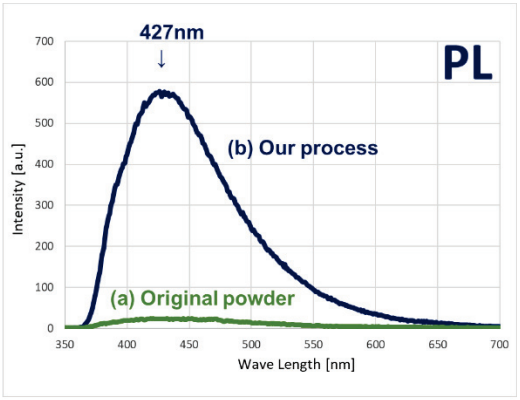


Fig. 3 PL spectra of InGaN/IPA solution of (a) original powder and (b) our process

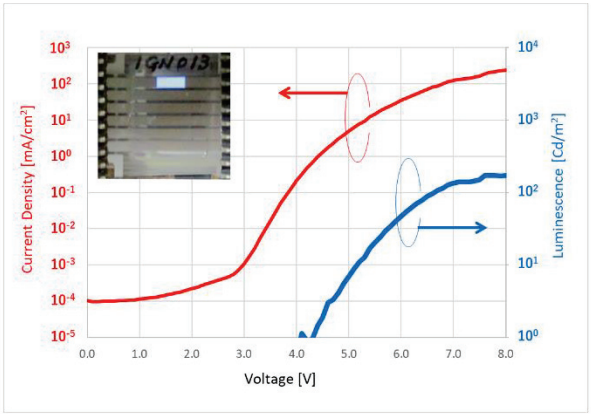


Fig. 4 V-I-L characteristics of solution processed InGaN LED

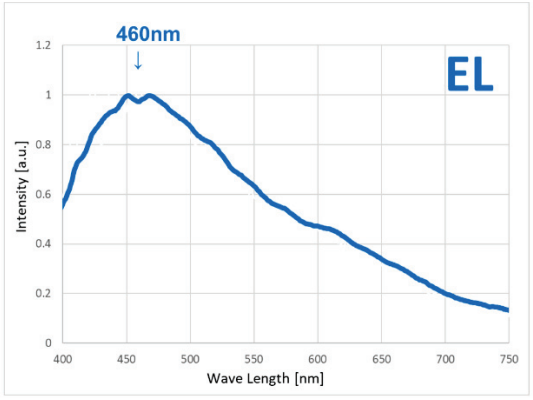


Fig. 5 EL spectrum of solution processed InGaN LED

Table 1 Device performance of the solution processed InGaN LED.

Current density [mA/cm^2]	Voltage [V]	Luminescence [Cd/m^2]	CIE_X	CIE_Y	L/I [Cd/A]
1	4.4	1.5	0.272	0.299	0.15
10	5.3	13.6	0.215	0.231	0.14

4 DISCUSSION

In this paper, we were able to fabricate a blue-emitting LED that incorporated an InGaN-based emissive layer which was fabricated by a solution processing method. The broad PL spectrum of the InGaN-containing solution indicates that optimization of the synthetic process is required to achieve an emission spectrum comparable to existing blue-emitters in the marketplace. Furthermore, the EL, V-I-L and performance of the devices indicates that additional improvements are required to the fabrication procedure to obtain a high power efficiency. Despite these shortcomings, we anticipate that, once optimized, these devices will have an enhanced lifetime in comparison to other organics and inorganic solution-processed blue emitters due to the inherent high stability of InGaN.

5 CONCLUSIONS

We developed a novel luminescent solution that contains an InGaN-based emissive material. This solution was used in the fabrication of the world-first blue LED that used a solution-processed InGaN-based emissive layer. With improvements to the preparation of this solution and the LEDs, it is anticipated that this route will become a competitor to existing, soluble organic and inorganic blue emitting materials.

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