



# Investigation on the wetting issues in solution processed inverted quantum-dot light-emitting diodes

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## ABSTRACT

Solution processed inverted quantum-dot light-emitting diodes (QLEDs) are developed in this work. To protect the QDs from dissolving by the solvents of upper layers, two commonly used hole transporting layer (HTL) materials for inverted QLEDs, poly (9-vinylcarbazole) (PVK) and 4-butylphenyl-diphenyl-amine (TPD), and three different solvents, chlorobenzene (CB), methylbenzene (MB) and chloroform (CF) are first investigated. The QDs layer has been less affected by CB solvent than another two solvents, meanwhile, we found that the device PL peak wavelength can be modulated by the HTL materials categories. Second, after overall estimating the wetting enhancement ability and the additive residue of several different additives, IPA was selected as additive to facilitate the deposition of a hydrophilic poly(ethylenedioxythiophene)/polystyrenesulfonate (PEDOT:PSS) hole injection layer on top of hydrophobic PVK. Finally, the best-performing device with improved electronic properties is achieved. The main findings in present work may offer a practicable strategy for further research, leading to achieve more efficient solution processed inverted QLEDs.

## 1. Introduction

Quantum-dot light-emitting diodes (QLEDs) are attracting considerable attention continuously as they are regarded as the potential candidate for next generation display and solid state lighting owing to their unique features, such as narrow band emission, size-tunable bandgaps, and high photoluminescence quantum efficiency [1–7]. The research in this field have grown rapidly and led to extraordinary progresses since the introduction of the first QLEDs [8–18]. Among these reported works, the numerous effects have been dedicated to optimizing the chemical synthesis of QDs or designing proper device architectures [12–18].

Inverted device structure rather than the conventional structure is becoming popular, as it is very advantageous for future QLEDs applications [12–14]. The bottom transparent cathode can be directly connected to n-channel metal oxide or amorphous silicon based thin-film transistor (TFT) backplanes. Furthermore, unlike the conventional structure, the first spin coated layer of ZnO nanoparticles (NPs) on cathode can go through a high annealing temperature without damaging other layers. With regard to fabrication, the QLEDs by solution processing methods such as spin-coating, inkjet printing is preferred, as it is suitable for the development of large-area, low-cost, and flexible devices, which offers considerable advantages compared with

conventional thermal evaporation method in high vacuum [19–24]. Therefore, only inverted QLEDs that are fabricated by solution processing method are considered here.

Choi et al. demonstrated the first all solution-processed inverted QLEDs, but the performance is very poor [25]. Afterwards, Jang et al. improved the device performance by using polyoxyethylene tridecyl ether (PTE) as a surfactant to facilitate the deposition of aqueous poly (ethylenedioxythiophene)/polystyrenesulfonate (PEDOT:PSS) on the poly(4-butylphenyl-diphenyl-amine) (pTPD) [16]. However, the demonstrated devices still performed unsatisfactory. Apparently, challenges facing all-solution processed inverted QLEDs still exist. For the first one, the solvent erosion of the HTL on the QDs emission layer always happens. Since QDs can be dispersed in common organic solvents used for HTL, it is easily damaged by the HTL layer coated above. Although it is proved that the solvent erosion can be avoided by inserting insulator between the QDs layer and HTL to defend against the solvent's attack, the driving voltage increases as the thickness of insulator inserted increase [26]. For another one, it is always difficult to deposit the aqueous PEDOT:PSS onto the underlying HTL layer, which is usually hydrophobic in nature.

In our contribution, to tackle the aforementioned issues in solution processed inverted QLEDs, the HTL materials and solvents utilized are first investigated. Two commonly used HTL materials for inverted

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QLEDs, poly (9-vinylcarbazole) (PVK) and 4-butylphenyl-diphenylamine (TPD) were dissolved in three solvents, chlorobenzene (CB), methylbenzene (MB) and chloroform (CF). Verified by the devices performances and the photoluminescence (PL) results, we found that QDs layer has been less affected by CB than the other two solvents. Meanwhile, it is observed that there is no shift of the PL peak wavelength after coating the QDs layer with PVK, while a blue shift appears after coating with TPD. Second, to deposit a uniform PEDOT:PSS layer, the wetting performance of PEDOT:PSS aqueous solution was improved by three additives, isopropanol (IPA), fluorocarbon surfactant (FC-4430), and dimethyl sulfoxide (DMSO) respectively. Although the FC-4430 performs best in the reduction of solution contact angle which means the wetting performance has been improved most significantly, the result of EDS indicates that there is nonnegligible residue in the PEDOT:PSS film, which will negatively affect the conductivity or hole injection efficiency. Finally, with optimized conditions, solution processed inverted QLEDs have been demonstrated. The results included in this contribution may offer a practicable strategy for further research, leading to achieve more efficient solution processed inverted QLEDs.

## 2. Experimental section

In order to fully evaluate the damage of the QDs layer caused by the HTL solvents, seven samples with structures ITO/ZnO/QDs, ITO/ZnO/QDs/PVK (dissolved in CF), ITO/ZnO/QDs/PVK (dissolved in CB), ITO/ZnO/QDs/PVK (dissolved in MB), ITO/ZnO/QDs/TPD (dissolved in CF), ITO/ZnO/QDs/TPD (dissolved in CB), and ITO/ZnO/QDs/TPD (dissolved in MB) were firstly fabricated, and the PL intensity of the QDs were compared. Afterwards, the wetting issue between PEDOT:PSS solutions and hydrophobic HTL was experimentally investigated. The role of three different additives, IPA, FC-4430, and DMSO in the improvement of wetting performance was successively investigated. Meanwhile, the additive residue was carefully inspected by EDS analysis. The final selection of additive for the following device fabrication is determined by comprehensively considering the wetting performance and the result of EDS analysis.

Then, for device performance investigation, the hole-only devices were the first to be fabricated and tested with the following structure: ITO/HTL/PEDOT:PSS (additive)/Al. For PEDOT:PSS solution, different additive concentrations and annealing temperature varied from 70 °C, 120 °C, and 200 °C were adopted to thoroughly examine the influencing factors and related mechanism of the wetting issues. Afterwards, solution processed inverted QLEDs with structures of ITO/ZnO/QDs/HTL/PEDOT:PSS(additive)/Al were fabricated. All layers (except for the Al electrode) were fabricated by spin-coating method under ambient air conditions. The ZnO film was firstly spin-coated at 2000 rpm onto ITO from a 30 mg/mL butanol solution, followed by baking at 200 °C for 30 min. Then, green emitting GA-CdSe/ZnS QDs purchased from Suzhou Kingshuo Nanotech Co., Ltd with emission wavelength at 528 nm were dissolved in n-octane (10 mg/mL) and then spin-coated at 1000 rpm for 30 s, followed by annealing at 120 °C for 10 min. The selected HTL material was spin-coated at 1000 rpm for 30 s, followed by baking at 120 °C for 10 min. After that, PEDOT:PSS with suitable additive for wetting improvement were spin-coated (1500 rpm 25s) and baked for 15min. Finally, the top Al cathode was deposited with an active device area of 4 mm<sup>2</sup>.

Contact angles were measured using a Phoenix 600 measurement system. The current-voltage (I–V) characteristics were measured with a Keithley-2400 source-meter unit. The luminance of the devices was calibrated using a Minolta luminance meter (LS-100). All measurements were conducted in air at room temperature.

## 3. Results and discussion

The structure and energy levels of the device are schematically shown in Fig. 1(a) and (b). ZnO nanoparticles were used as electron

transport material due to their high electron mobility and matched energy level with that of QDs. The QDs dissolved in n-octane can be spin-coated onto ZnO without damaging the ZnO layer [6]. However, the QDs layer is easily dissolved by the solvents of HTL. To minimize the damage and maintain the excellent performance of QDs layer, the solvents of HTL and the materials itself should be carefully chosen. To this end, typical organic solvents CF, CB, and MB were investigated first. The samples were fabricated according to the structures introduced in Experimental section.

Fig. 2 shows the PL intensity and the photographs of these samples illuminated by a 410 nm ultraviolet light source. Samples without the HTL layer exhibited the highest PL intensity as the QDs remain intact. However, by spin-coating PVK or TPD onto QDs, the PL intensity has been decreased dramatically. Please note that, the decrease in PL intensity might be partially due to the absorption by the HTL materials, however, for present study, we are focusing on the solvent erosion issue of the HTL on the QDs emission layer. As reflected in the PL intensity, the QDs layer has been less affected by CB solvent than by MB and CF solvents for both PVK and TPD. This has also been verified by the photographs of the samples as displayed in inset of Fig. 2. It can be clearly observed that the center of the samples is relatively dark especially for the samples with MB and CF solvents, as the QDs in that region is severely dissolved by the solvents. Samples with CB solvent exhibited relatively higher PL intensity and more uniform emission, indicating that the damage caused by the CB is the smallest and thus CB is the best solvent for PVK and TPD.

Just as mentioned previously, the QDs can be well dissolved in all three above mentioned solvents CB, CF and MB, therefore, we believed that the reduced PL intensity is mainly due to the reduced thickness of the QDs layer. The solvents will penetrate into the QDs layer and wash out some QDs right after spin coating the PVK or TPD solution onto the QDs layer, which will finally lead to a reduced QDs thickness. To verify our assertion, we dropped the solvents onto the QDs layer and then spin-coating the solvents, the QDs thickness was then measured after baking to examine if it is affected by the solvents. The results indicated that the thickness of QDs layer was reduced from 30 to 23 nm, 19 and 13 nm, when the CB, MB and CF were respectively spin-coated onto the QDs layer. Such results could be further well explained by the solvents viscosity, which actually affects the penetration rate of solvents into QDs. Since the viscosity of CB is the largest, the penetration rate of CB is thus the slowest, and as a result, the damage on QDs layer thickness is the slightest. Meanwhile, we also investigated the influence of HTL materials, e.g. PVK and TPD on the PL intensity of samples. We interestingly found that there is a blue shift of the PL peak wavelength after coating the QDs layer with TPD, which has also been verified by the photographs of the samples as displayed in inset of Fig. 2. It can be clearly observed that there is only one PL peak wavelength for the case of PVK, while the samples with TPD coating exhibit a second peak in the relatively high-frequency band region. Since the PVK coating is usually transparent, the photographs of samples coated with PVK exhibits the same green color as the sample without the HTL layer. However, as the color of TPD solution is pale yellow, therefore, the detected PL peak wavelength is somehow modulated and overall exhibits a blue color as shown in the photographs. Although such modulation in PL peak wavelength may be useful in some specific applications, the main task of present study is taking fundamental investigation of optoelectronic properties of solution processed inverted QLEDs. Therefore, PVK and CB were selected as the HTL material and the solvent respectively, for the following device performance investigations.

Another challenge of making solution processed inverted QLEDs is the wetting issue between adjacent layers, especially for the coating process of the aqueous PEDOT:PSS onto the PVK layer, which is hydrophobic in nature. To overcome this difficulty, additive could be used to facilitate the deposition of PEDOT:PSS onto PVK layer. The effect of three different additives, IPA, FC-4430 and DMSO on the wetting

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