

RESEARCH ARTICLE

Inkjet-printed multi-color arrays based on eco-friendly quantum dot light emitting diodes with tailored hole transport layer

Shijie Zhan  | Yo-Han Suh | Xiang-Bing Fan | Jiajie Yang | Limeng Ni | Yoonwoo Kim | Jeong-Wan Jo | Hyung Woo Choi | Sung-Min Jung | Dong-Wook Shin | Sanghyo Lee  | Jong Min Kim

Department of Electrical Engineering,
University of Cambridge, Cambridge, UK

Correspondence

Dong-Wook Shin and Sanghyo Lee,
Department of Electrical Engineering,
University of Cambridge, 9 JJ Thomson
Avenue, Cambridge CB3 0FA, UK.
Email: ds887@cam.ac.uk and
sl920@cam.ac.uk

Funding information

Engineering and Physical Sciences
Research Council, Grant/Award Number:
EP/P027628/1; H2020 Industrial
Leadership, Grant/Award Number:
685758

Abstract

Cadmium-free quantum dot light-emitting diodes (QLEDs) have held the potential to revolutionize the next-generation displays with their advantages in color gamut, luminance intensity, and solution processibility. As a promising way of realizing large-area QLED display production, inkjet printing has been intensively studied on Cd-based QLEDs but lacks exploration in fabricating Cd-free devices. Here, we developed Cd-free RGB inkjet-printed QLEDs with tailored hole transport layers (t-HTLs) using Cd-free QDs including InP/ZnSeS red and green QDs and ZnTeSe/ZnSe/ZnS blue QDs. With the t-HTLs, QD ink erosion on the bottom charge transport layer was remarkably suppressed, while the efficient hole transport was maintained, which kept high device performance, especially in the QLED lifetime. With bank structures, Cd-free QLED pixels were well defined within the size of $60\ \mu\text{m} \times 160\ \mu\text{m}$. Based on the t-HTL structure and the bank structures, inkjet-printed Cd-free RGB QLED pixel arrays were demonstrated. This study bridges the gap between existing Cd-free QLED technologies and the future commercialization of Cd-free self-emissive QD displays.

KEYWORDS

bank structure, cd-free RGB QDs, inkjet printing, QLEDs, tailored hole transport layer

1 | INTRODUCTION

Quantum dot light-emitting diodes (QLEDs) with the unique advantages of quantum dots (QDs) in size-tunable emission wavelength, high color purity, and cheap solution-based processibility¹ have been rapidly developed for future self-emissive display technology. To realize the display with RGB QLEDs, a cost-effective manufacturing method with pixelated RGB QD patterns

is required. Among numerous techniques, the inkjet printing method has been regarded as the most promising large-area fabrication method to realize self-emissive QLED displays with a near-unity material utilization rate and a low-cost manner.²

Up to date, a variety of studies have been conducted on inkjet-printed Cd-based QLEDs, including solvent mixture, ligand exchange, and transport layer modification.³ To achieve high pixel resolution and uniformity of QD

This is an open access article under the terms of the [Creative Commons Attribution](https://creativecommons.org/licenses/by/4.0/) License, which permits use, distribution and reproduction in any medium, provided the original work is properly cited.

© 2022 The Authors. *Journal of the Society for Information Display* published by Wiley Periodicals LLC on behalf of Society for Information Display.

films using inkjet printing method, cyclohexylbenzene (CHB) as the solvent for QD ink formulation was mixed with an additional solvent, such as octane, nonane, and dichlorobenzene, resulting in the prevention of coffee ring effect.^{3a-c} Besides, electrohydrodynamic printing and dual ion passivation of Cd-based QDs by zinc oleate have been adopted to achieve high-resolution Cd-based QLED pixel arrays^{3c} and high device performance,^{3d} respectively. In addition, to avoid the solubility issue of the bottom hole transport layer during ink-jetting, inverted QLED structures have been widely employed because of the high ink resistance of ZnO nanoparticle films as electron transport layers (ETLs).^{3a,e}

However, due to the intrinsic toxicity of Cd-based QDs, breakthroughs on a QD material itself for the inkjet printing method are required. Recently, Cd-free QDs (eco-friendly) have been significantly improved so that high QLED performance was achieved (spin-coated Cd-free red InP and blue ZnTeSe QLEDs with both EQE over 20%⁴), but there is still a lack of research about inkjet-printed RGB Cd-free QLEDs with pixel arrays. In addition, it is necessary to understand interfaces between QD and charge transport layer, especially polymeric hole transport layer (HTL), in the conventional QLED structure that has the state-of-the-art of QLED performance.^{4,5} Therefore, to realize the display technology based on Cd-free QLEDs fabricated by inkjet printing, it is crucial to understand jetting behavior of ink containing InP and/or ZnTeSe-based QDs and investigate the interfaces between QD and transport layer in the device.

Here, we demonstrated eco-friendly inkjet-printed RGB QLED arrays based on a conventional device structure with a tailored hole transport layer overcoming an erosion by QD ink droplets. An optimized QD ink was developed using a mixture of cyclohexylbenzene and octane. To prevent the erosion of ink on the bottom HTL, a tailored double HTL was adopted to fabricate InP red, InP green, and ZnTeSe blue-based QLEDs without parasitic emission from HTL. The surface of tailored HTL resisted the printed ink and enabled higher device performance and a longer device lifetime. Based on its structure, inkjet-printed RGB Cd-free QLED arrays with bank structures were fabricated. This study will pave the way to realize next-generation self-emissive displays with eco-friendly QLEDs in the future.

2 | RESULTS AND DISCUSSION

2.1 | QD ink and printing optimization

For Cd-free QD materials used in inkjet printing, InP/ZnSeS QDs (InP core and ZnSeS gradient shell) were

chosen for red and green emission. Due to the nature of the small bandgap in bulk InP (1.344 eV), well-synthesized InP QDs could only emit cyan or light blue color with a peak wavelength longer than 460 nm.^{4b,6} Therefore, ZnTeSe/ZnSeS/ZnS QDs were selected for blue emission because of the large bulk bandgap of ZnSe and the high efficiency through gradient shell coating.^{4b,7} Figure 1A–C shows the photoluminescence (PL) and absorption spectra of red, green, and blue Cd-free QDs. The PL peaks of InP/ZnSeS red, InP/ZnSeS green, and ZnTeSe/ZnSe/ZnS maximize at 605 nm (FWHM, 55.34 nm), 532.6 nm (39.25 nm), and 442.2 nm (45.35 nm), respectively.

Figure 1D shows the diagram of QD inkjet printing, where QD ink was printed onto HTL. To print the RGB QD inks, the control waveform for jetting ink droplet was set into a five-stage process, as shown in Figure S1 (see Methods). The stable jetting conditions, such as solvent and temperature, of green InP-based QD ink were investigated, as shown in Figure S2 (see “Comparison of Dodecane and CHB QD Ink” in the supporting information). Cyclohexylbenzene (CHB) is used as the primary solvent of ink formulation for Cd-free RGB QDs, together with the processing temperature on the substrate at 50°C, as considered the film uniformity and dimension, and droplet conditions during printing. It is noted that CHB QD ink was able to eject without nozzle blocking (Figure S3A), based on the optimized waveform (Figure S1) while the dodecane based QD ink was blocked nozzle easily. By calculating the displacement of the drops within 0.5 s, the average velocity of the drops was ~304 μm/s (Figure S3B).

However, dots printed from droplets of CHB ink still suffered from the coffee-ring effect to some degree. One of the strategies to suppress the outward flow of QD solutes during droplet evaporation is to mix the solvent with another solvent with lower surface tension, resulting in the prevention of the coffee-ring effect due to inward flow based on the Marangoni effect.^{3a} Specifically, liquid at the edge part of a droplet will evaporate faster, leaving more CHB, compared to the central part of the droplet, which can induce inward flow (Marangoni flow) as shown in Figure 1E. Octane with a surface tension of 21.6 mN/m was selected as the additional solvent, while the surface tension of CHB is 36.3 mN/m.^{3b,8} Different ratios of octane (0 to 5%) were added into CHB QD ink for comparison (Figures 1F and S4, PL images of dot arrays at substrate temperatures, 25°C and 50°C).

At 25°C (Figure 1F, i–iii), regardless of volume percent of octane, droplets printed have a coffee-ring effect, which was confirmed by the line profile of PL image (Figure S5A–C). By increasing the substrate to 50°C, due to fast evaporation of the solvent, balanced inward and

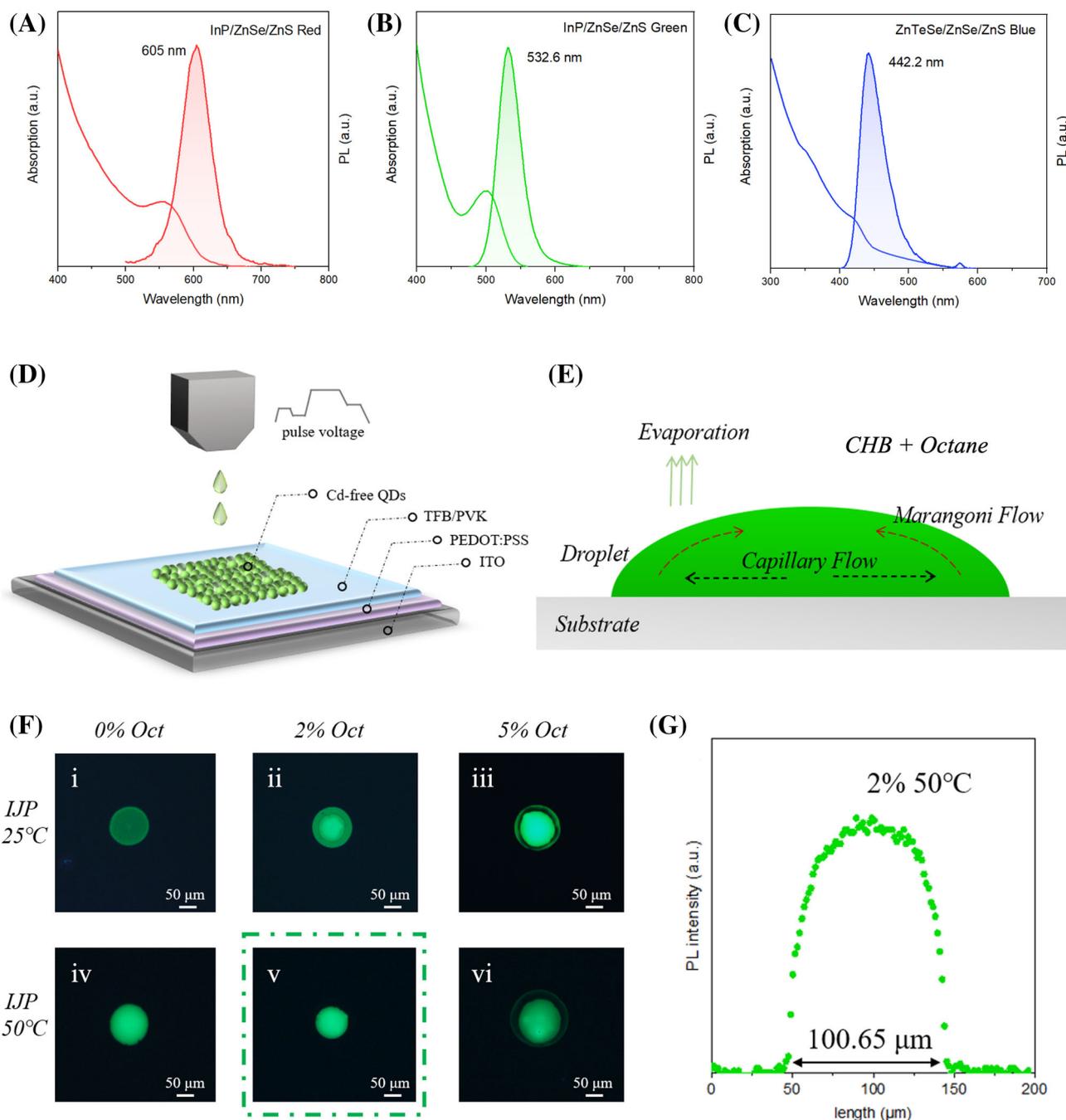


FIGURE 1 RGB eco-friendly QDs and optimization of inkjet printing. (A–C) The PL and absorption spectra of Cd-free QDs used for fabrication of QLED by inkjet printing method, including (A) InP/ZnSeS red, (B) InP/ZnSeS green, and (C) ZnTeSe/ZnSeS/ZnS blue. (D) Schematics of the QLED inkjet printing process. (E) Schematic illustration of drying process of a droplet. (F) Enlarged PL images of printed dots (CHB based ink with various volume ratios of octane, 0, 2, and 5%): At 25°C, (i) 0%, (ii) 2%, and (iii) 5% of octane; At 50°C, (iv) 0%, (v) 2%, and (vi) 5% of octane. The best condition was reached with 2% octane and substrate temperature of 50°C. (G) The line profile of PL intensity across the dot printed with CHB ink containing 2% octane at the substrate temperature of 50°C

outward flows were achieved. As the octane ratio is 0% or 2% (Figure 1F, iv and v), uniform PL profile of the printed dot was confirmed without any bumps and glitches (Figure S5D,E). In particular, the dot printed with ink containing 2% octane and substrate temperature at 50°C had the minimum diameter (100.65 μm) and

most uniform PL distribution (Figure 1G). It was noticed that, with 5% octane, the center and edge of printed droplets became separated, which indicated the Marangoni effect became excessive (Figure 1F, iii and vi). Compared to the ink without any octane addition, the CHB-based ink with 2% octane enabled a smaller dot pattern size

because of the optimized Marangoni inward flow. Therefore, printing CHB based ink with 2% octane onto 50°C substrate was adopted in the following device fabrication section.

2.2 | Ink erosion on HTL

This work employs a conventional QLED structure (Figure S6), anode/hole injection layer (HIL)/HTL/QDs/electron transport layer (ETL)/cathode, where indium tin oxide (ITO), poly-(ethylenedioxythiophene):polystyrenesulfonate (PEDOT:PSS), ZnMgO nanoparticles, and Al are the anode, HIL, ETL, and cathode, respectively. Poly(9,9-dioctylfluorene-alt-N-[4-sec-butylphenyl]-diphenylamine) (TFB) or poly(9-vinylcarbazole) (PVK) were used for HTL. Cd-free QDs ink in CBH/octane, including red and green InP-based QDs and blue ZnTeSe based QDs, were inkjet-printed with the optimized conditions (see Appendix A). The thickness of each layer was measured by a DektakXT profiler system (Figure S7).

One of the critical issues in the conventional device structure is the erosion effect of HTL during inkjet printing of QD ink, which affects device performance. The CHB/octane QD ink could cause dissolving bottom HTL and forming an eroded QD/HTL interface as depicted in Figure 2A.^{4a,5,9} Within the eroded area, the carriers could transport through the leakage paths due to direct contact between both charge transport layers, resulting in the decrease of the device efficiency and light emission. Figure S8 shows the vulnerability of TFB upon CHB/octane QD ink, while PVK possesses better ink resistance (see “Ink Erosion on HTL” in the supporting information). The contact angle of the mixed solvent (CHB with 2% octane) on different HTL materials was further measured (Figure 2B), showing that the contact angles of mixed solvent on a quartz coated glass, a TFB film, and a PVK film were 12.6°, 9.3°, and 6.1°, respectively. The smaller contact angle of the ink solvent on PVK resulted in better ink wetting during QD printing, compared to the wetting on TFB. Both the stronger resistance and better ink wetting make PVK a suitable HTL for inkjet-printed QLEDs.

To further investigate the ink erosion for inkjet-printed QLED performance, electroluminescence (EL) of inkjet-printed green InP/ZnSeS QLEDs was analyzed with various fabrication conditions including different HTL materials, HTL annealing conditions, and substrate temperature (Figure 2C–H). Since the printing process was conducted in an ambient condition, the substrate temperature was limited to 50°C to avoid severe QD degradation. It was noted that, at the substrate temperature of 25°C and 50°C, the printed QLED with TFB annealed

at 150°C had EL spectrum dominant by TFB emission at 435 nm and QD/TFB interlayer emission ~630 nm instead of green emission, which corresponds to the exciplex recombination from the conduction band in QDs to the valance band in TFB (Figure 2C).¹⁰ At the substrate temperature of 25°C, printed QLED with TFB annealed at 200°C still had a dominant TFB emission with small green QD emission in the EL spectrum (Figure 2D). It reveals that there is still a violent intermix of QD and TFB before film drying, which could lead to exciton recombination in the TFB film. With the substrate temperature rising to 50°C, printed QLEDs had a green QD EL spectrum with relatively small TFB emission (Figure 2E). This is attributed to higher substrate temperature that facilitates solvent evaporation of droplets and simultaneously reduces ink erosion time.¹¹ However, high annealing and high substrate temperature still could not fully get rid of the parasitic TFB emission, leaving the pixel with a bluish-green color. As Figure 2F,E presents, the printed QLED with PVK, even at lower HTL annealing (150°C) or substrate temperature (25°C), had a strong green emission with a small parasitic PVK emission at ~415 nm, indicating the higher endurance of PVK to ink erosion effect compared with TFB. With PVK annealed at 200°C and substrate temperature at 50°C, a green EL spectrum without blue PVK parasitic emission was achieved. It is noteworthy that parasitic emission from HTLs can induce HTL and device degradation and result in bluish emission, which must be eliminated to achieve efficient QLED devices.¹²

2.3 | Ink-erosion-free Cd-free printed QLEDs with tailored HTL structures

Though PVK has superior solvent resistance, its carrier mobility ($2.5 \times 10^{-6} \text{ cm}^2/\text{Vs}$) is much lower than that of TFB ($2 \times 10^{-3} \text{ cm}^2/\text{Vs}$), which causes inferior QLED performance.¹³ To combine the advantage of TFB and PVK, a tailored double HTL structure of TFB/PVK was adopted in this work. Considering the large thickness of the double HTL, the TFB bottom layer was partially removed by a chlorobenzene casting technique as shown in Figure S9A, where a drop of chlorobenzene was casted on a spinning film.¹⁴ To compare the effect of tailored double structure with tailored-TFB/PVK (t-TFB/PVK), spin-coating method for QD layer deposition was initially employed to confirm the device performance without ink erosion effect based on HTL of PVK, TFB/PVK, and t-TFB/PVK, as shown in Figure S9. QLED with t-TFB/PVK shows the highest efficiency while maintaining high luminance. The thickness of t-TFB (6.4 nm), t-TFB/PVK (19.1 nm), and TFB/PVK (27.4 nm) was obtained

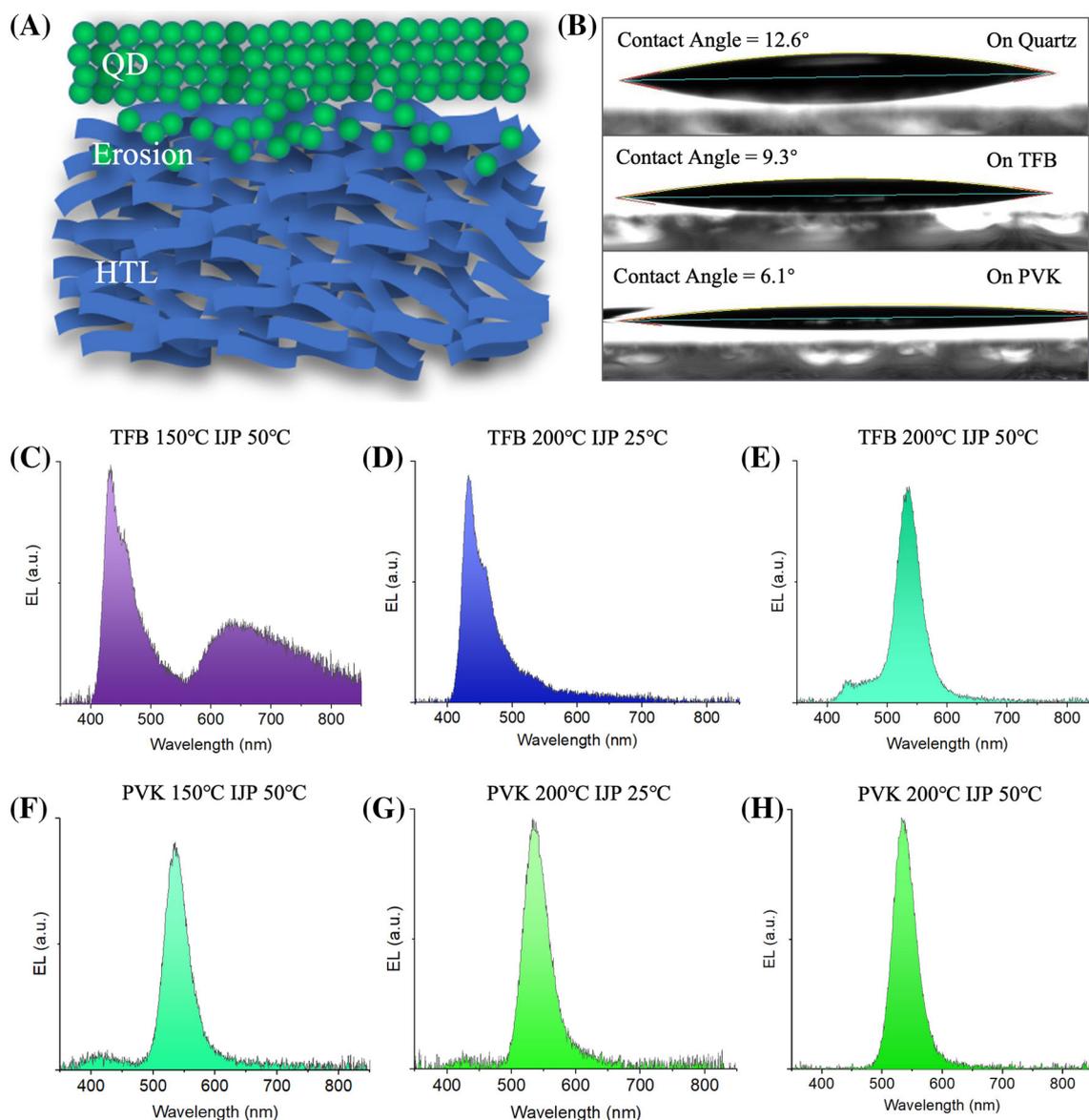


FIGURE 2 Ink erosion of HTL. (A) Schematics of the QD ink erosion on an HTL layer. (B) The contact angle of CHB/octane (2%) on quartz, TFB film, and PVK film. (C–H) The EL spectra of inkjet-printed green InP/ZnSe QLEDs based on HTL of TFB with annealing and substrate temperature at (C) 150°C and 50°C, (D) 200°C and 25°C, (E) 200°C and 50°C; and the EL spectra of QLEDs based on HTL of PVK with annealing and substrate temperature at (F) 150°C and 50°C, (G) 200°C and 25°C, and (H) 200°C and 50°C

(Figure S10). We observed that, with the chlorobenzene spin-casting process (Figure S9A), the thickness of TFB was reduced from 22.3 to 6.4 nm.

To realize the small size of RGB pixel dimension for next-generation display with self-emissive QLED using inkjet printing technology, we fabricated bank arrays by a conventional photolithography.¹⁵ Photoresist bank arrays were fabricated on ITO substrates before PEDOT:PSS coating, as pixel-defined substrates (see Appendix A.3, 20 μm vertical gaps and 40 μm horizontal gaps between pixels with the pixel size of 60 μm \times 160 μm , Figures S11–S13). With bank structures, the effectiveness

of tailored double HTL structures in inkjet-printed QLEDs was validated using green InP/ZnSe QDs ink with CHB/octane (2%). Figure 3A shows the schematic illustration of inkjet-printed QLEDs in bank structures that allow printed QD ink to locate into each pixel without cross-spreading. The EL images and performance of printed green QLED pixels in bank structures with different HTL structures are displayed, including TFB, PVK, TFB/PVK, and t-TFB/PVK (Figure 3B). Due to the ink erosion effect on TFB, the TFB-based printed QLEDs had bluish emission because of the parasitic emission from TFB (Figure 3B). In contrast with that, the PVK-based

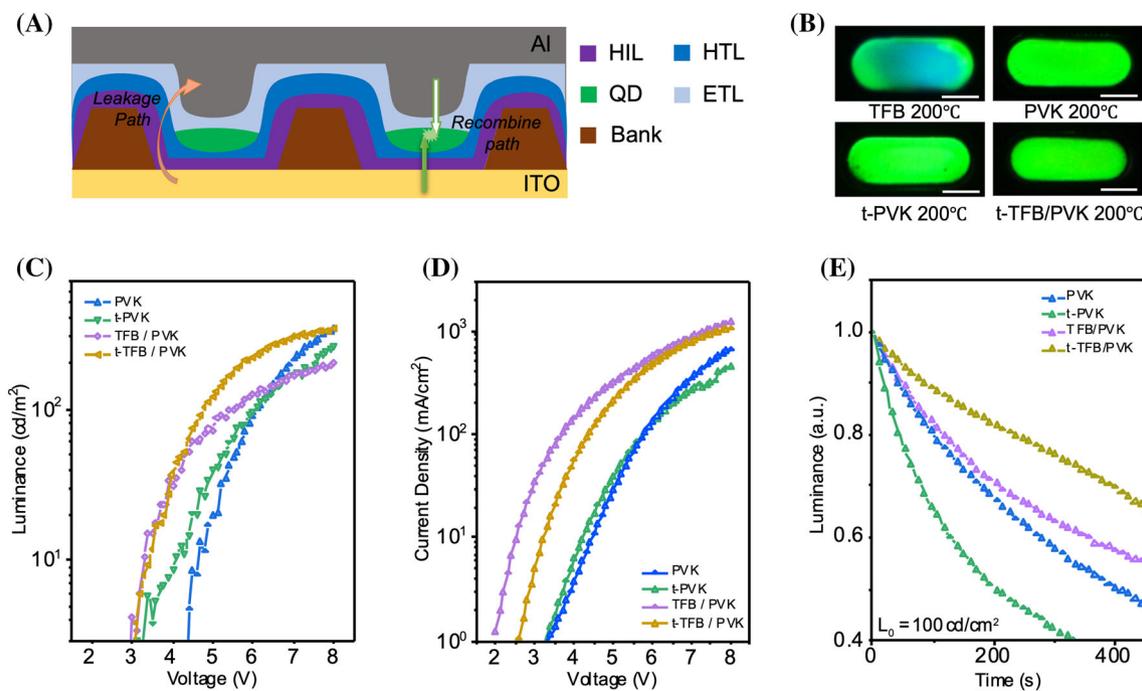


FIGURE 3 Inkjet-printed QLED with band structure. (A) Schematic illustration of inkjet-printed QLEDs with bank structures. (B) The EL images of green InP-based QLED pixels with different HTLs. Performance of QLEDs with different HTLs, including (C) the luminance-voltage and (D) the current density-voltage. (E) The lifetime of devices with different HTLs. The initial luminance of all devices was set to 100 cd/m^2

printed QLEDs had no bluish emission and possessed relatively high EQE, even if they suffer from high turn-on voltage due to a low hole transport mobility as discussed above. To enhance the device performance, the double layer HTL was applied to the inkjet-printed QLED. Compared to the PVK (or tailored PVK, t-PVK) and TFB/PVK based printed QLED, the device with t-TFB/PVK showed maximized luminance.

The double HTL structure can also expand the lifetime of inkjet-printed QLEDs (Figure 3E). InP green-based QLEDs with different HTLs were biased with constant current density (100 cd/cm^2) for lifetime measurement. The longest lifetime was obtained from t-TFB/PVK QLED among all inkjet-printed devices. In principle, the imbalance of electron/hole injection is the main reason for a short lifetime, which is caused by poor hole injection.¹⁶ In this case, the additional thin TFB layer between PVK and PEDOT:PSS increases the effective hole mobility, resulting in a more balanced charge injection so as to lead to a longer device lifetime, compared to the lifetime of the QLED with PVK. Compared to the PVK QLED with a T_{90} of $\sim 43 \text{ s}$, the t-TFB/PVK QLED had a doubled T_{90} lifetime. Based on the tailored double HTL structure (t-TFB/PVK), red/green/blue pixelated QLED arrays were fabricated by inkjet printing with red/green InP/ZnSeS

QDs and blue ZnTeSe/ZnSeS/ZnS QDs (Figure S14). It is noteworthy that relatively lower lifetime of inkjet-printed QLED with the eco-friendly QDs, especially InP based QDs, stems from the weak bonding between In and P in air. To further improve the lifetime of inkjet printed QLEDs, studies on protecting the surface of QDs in ambient conditions will be required.

With the bank structures, eco-friendly red/green/blue inkjet-printed QLED arrays were achieved. Figure 4 shows the EL spectra and pixel array images for red, green, and blue QLEDs fabricated by inkjet printing, with the tailored double HTL structures. The peak EL wavelength of red, green, and blue QLEDs was 612.5, 533.4, 445.5 nm, respectively (Figure 4A–C), which is confirmed by the CIE chromaticity coordinate at (0.628, 0.370), (0.308, 0.661), and (0.145, 0.084) (Figure S15). At different operating voltages, the emission wavelength did not show a significant shift. With the tailored double HTL structures, inkjet-printed red, green, and blue pixelated Cd-free QLED arrays were realized on the bank structures (Figure 4D–F). For a demonstration of Cd-free RGB inkjet-printed pixelated-QLED, arrays arranged alternatively with red, green, blue QLEDs were fabricated (Figure 4G,H). Figure 4I,J shows the whole images of printed pixel arrays with red, green, and blue light emission on various platforms.

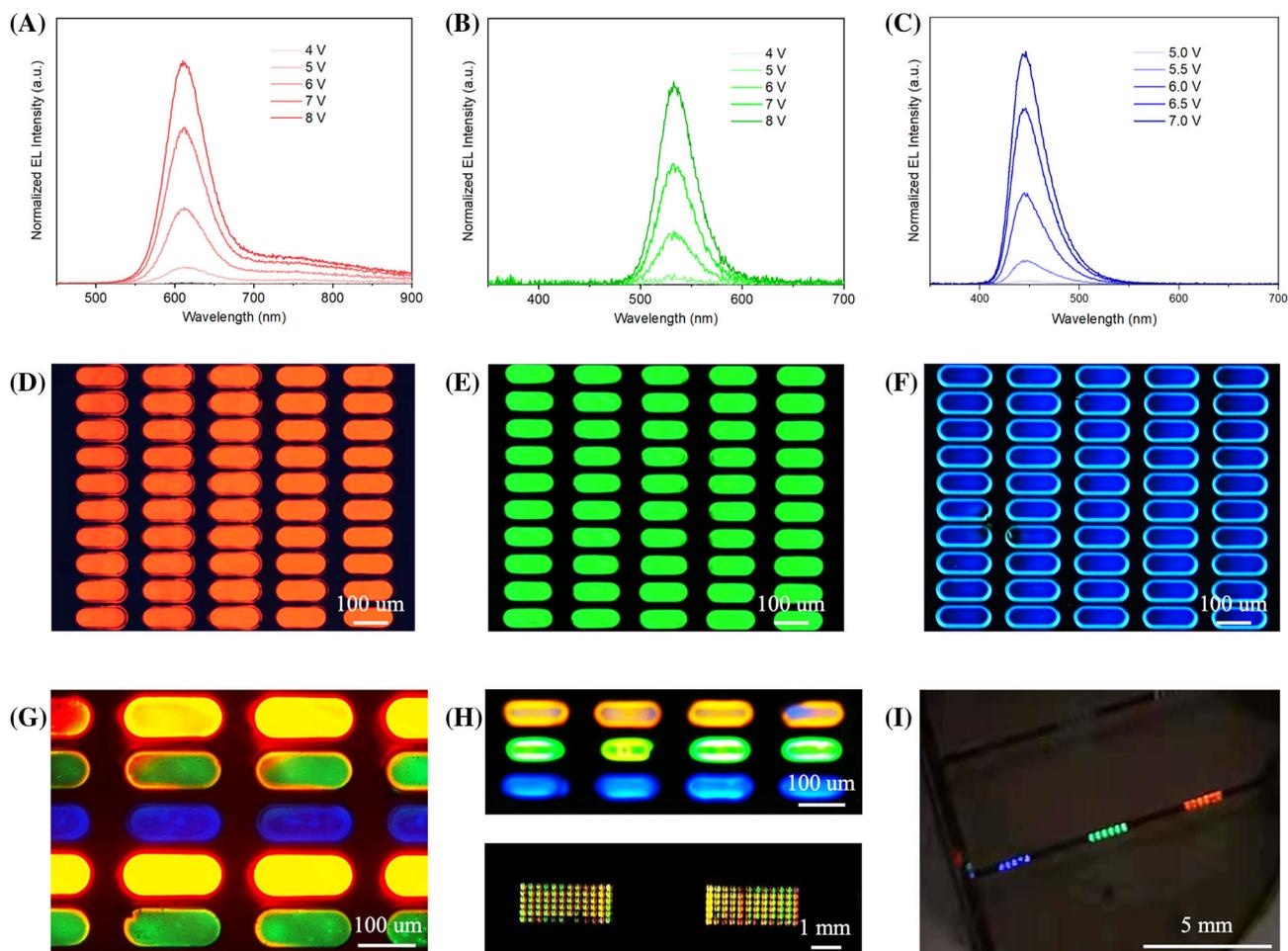


FIGURE 4 Eco-friendly RGB QLED arrays. (A–C) The EL spectra of (A) InP/ZnSe red, (B) InP/ZnSe green, and (C) ZnTeSe/ZnSe/ZnS blue inkjet-printed Cd-free QLEDs with t-TFB/PVK. (D–F) The EL microscopic images of Cd-free QLED pixel arrays for (D) InP/ZnSe red, (E) InP/ZnSe green, and (F) ZnTeSe/ZnSe/ZnS blue. (G–I) The microscopic EL Images of Cd-free multi-color (red, green, and blue) QLED pixel arrays. (J) The EL image of InP/ZnSe red, InP/ZnSe green, and ZnTeSe/ZnSe/ZnS blue Cd-free QLEDs printed separately on the same substrate. All the devices were fabricated based on the tailored double HTL structures

3 | CONCLUSION

In this work, eco-friendly RGB QLED arrays were for the first time achieved by inkjet printing. By designing tailored double HTL structures, ink erosion on HTL can be remarkably suppressed, which improves device performance. InP/ZnSeS red and green and ZnTeSe/ZnSeS/ZnS blue QDs ink in CHB mix with 2% octane were adopted for inkjet printing, which can achieve a minimum droplet diameter ~ 50 μm at the substrate temperature of 50°C. To achieve high-performed QLEDs fabricated by inkjet printing, a tailored double HTL was proposed with a PVK layer deposited on a solvent-rinsed TFB layer, which provided resistance to the ink and sufficient hole-transport property. Based on this HTL structure, red, green, and blue inkjet-printed Cd-free QLEDs were achieved low turn-on

voltage and high luminance. With photoresist banks fabricated on ITO substrates, high-resolution RGB color QLED pixels down to 60 μm × 160 μm were demonstrated. This work paves the way for the development of Cd-free electroluminescent QLED for future display applications.

ACKNOWLEDGMENTS

This work was supported by the European Commission (H2020 Industrial Leadership, 1D-NEON, Grant agreement ID: 685758) and the UK Research and Innovation (Engineering and Physical Sciences Research Council, EP/P027628/1).

ORCID

Shijie Zhan  <https://orcid.org/0000-0002-7566-6072>

Sanghyo Lee  <https://orcid.org/0000-0002-4723-9612>

REFERENCES

- Dai X, Deng Y, Peng X, Jin Y. *Adv Mater*. 2017;29(14):1607022. <https://doi.org/10.1002/adma.201607022>
- Lan L, Zou J, Jiang C, Liu B, Wang L, Peng J. *Front Optoelectron*. 2017;10(4):329–52. <https://doi.org/10.1007/s12200-017-0765-x>
- a) Jiang C, Zhong Z, Liu B, He Z, Zou J, Wang L, et al. *ACS Appl Mater Interfaces*. 2016;8(39):26162–26168. <https://doi.org/10.1021/acsami.6b08679>; b) Yang P, Zhang L, Kang DJ, Strahl R, Kraus T. *Adv Opt Mater*. 2020;8(1):1901429. <https://doi.org/10.1002/adom.201901429>; c) Li H, Duan Y, Shao Z, Zhang G, Li H, Huang Y, et al. *Adv Mater Technol*. 2020;5(10):2000401. <https://doi.org/10.1002/admt.202000401>; d) Xiang C, Wu L, Lu Z, Li M, Wen Y, Yang Y, et al. *Nat Commun*. 2020;11(1):1; e) Han J, Ko D, Park M, Roh J, Jung H, Lee Y, et al. *J Soc Inf Disp*. 2016;24(9):545–51. <https://doi.org/10.1002/jsid.467>
- a) Won Y-H, Cho O, Kim T, Chung D-Y, Kim T, Chung H, et al. *Nature*. 2019;575(7784):634–8. <https://doi.org/10.1038/s41586-019-1771-5>; b) Kim T, Kim K-H, Kim S, Choi S-M, Jang H, Seo H-K, et al. *Nature*. 2020;586(7829):385–9. <https://doi.org/10.1038/s41586-020-2791-x>
- H. Shen, Gao Q, Zhang Y, Lin Y, Lin Q, Li Z, et al. *Nat Photonics*. 2019;13(3):192–7. <https://doi.org/10.1038/s41566-019-0364-z>
- Ippen C, Guo W, Zehnder D, Kim D, Manders J, Barrera D, et al. *J Soc Inf Disp*. 2019;27(6):338–46. <https://doi.org/10.1002/jsid.780>
- Jang E-P, Han C-Y, Lim S-W, Jo J-H, Jo D-Y, Lee S-H, et al. *ACS Appl Mater Interfaces*. 2019;11(49):46062–46069. <https://doi.org/10.1021/acsami.9b14763>
- Chhatre SS, Guardado JO, Moore BM, Haddad TS, Mabry JM, McKinley GH, et al. *ACS Appl Mater Interfaces*. 2010;2(12):3544–54. <https://doi.org/10.1021/am100729j>
- Jang E, Kim Y, Won Y-H, Jang H, Choi S-M. *ACS Energy Lett*. 2020;5(4):1316–27. <https://doi.org/10.1021/acscenergylett.9b02851>
- Kim H-B, Kim J-J. *J Inf Disp*. 2019;20(3):105–21. <https://doi.org/10.1080/15980316.2019.1650838>
- Kim HG, Lee SM, Shin D, Ha J, Lee C. *SID Symp Dig Tech Papers Wiley Online Library*. 2020;51:395–7. <https://doi.org/10.1002/sdtp.13887>
- a) Kim BH, Acharya KP, Titov A, Zhang X, Holloway P. *SID Symp Dig Tech Papers Wiley Online Library*. 2021;147–50; b) Wu Z, Zhang W, Liu P, Wang K, X W. Sun. *SID Symp Dig Tech Papers Wiley Online Library*. 2020;754–7.
- a) Lin Q, Shen H, Wang H, WanG A, Niu J, Qian L, et al. *Org Electron*. 2015;25(178):183. <https://doi.org/10.1016/j.orgel.2015.06.032>; b) Pan J, Chen J, Huang Q, Wang L, Lei W. *RSC Adv*. 2017;7(69):43366–43372. <https://doi.org/10.1039/C7RA08302E> c) Zhao Y, Chen L, Wu J, Tan X, Xiong Z, Lei Y. *IEEE Electron Dev Lett*. 2019;41(1):80–3. <https://doi.org/10.1109/LED.2019.2953088>
- Liu Y, Cui J, Du K, Tian H, He Z, Zhou Q, et al. *Nat Photonics*. 2019;13(11):760–4. <https://doi.org/10.1038/s41566-019-0505-4>
- Ko D, Han J, Roh H, Park Y, Lee J, Kim J, et al. *SID Symp Dig Tech Papers Wiley Online Library*. 2018;49:1629–31. <https://doi.org/10.1002/sdtp.12294>
- Liu D, Cao S, Wang S, Wang H, Dai W, Zou B, et al. *J Phys Chem Lett*. 2020;11(8):3111–5. <https://doi.org/10.1021/acs.jpcclett.0c00836>

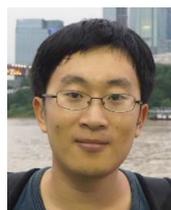
AUTHOR BIOGRAPHIES



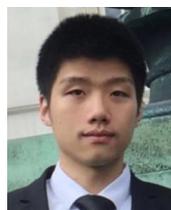
Shijie Zhan is currently working at Huawei Hisilicon Research Department as a senior research engineer. He received his PhD from the Department of Engineering, University of Cambridge, in 2021. He received his Bachelor's Degree from Zhejiang University in 2017.



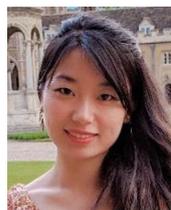
Yo-Han Suh worked at the Department of Engineering, University of Cambridge, as a research associate from 2018 to 2021. He received a PhD in the School of Energy and Chemical Engineering, Ulsan National Institute of Science and Technology (UNIST) in 2018.



Xiangbing Fan is currently working at the Department of Engineering, University of Cambridge, as a research associate. He received his PhD from the Technical Institute of Physics and Chemistry, Chinese Academy of Science, in 2018.



Jiajie Yang is currently a PhD Student at the University of Cambridge. He is working in the areas of smart textiles and quantum dot displays. He has a background in both electrical engineering (MPhil) and mechanical engineering (BEng).



Limeng Ni is currently working as a research associate at the Electrical Engineering Division, University of Cambridge. She was a Jardine scholar and received her PhD in Physics from Cambridge University.



Yoonwoo Kim is currently a PhD student in the Electrical Engineering Division, the Department of Engineering, University of Cambridge. He has completed his BS degree in the department of electronic engineering in 2019.



Jeong-Wan Jo is a postdoctoral researcher of the Department of Engineering, University of Cambridge. He received a PhD in the School of Electrical and Electronics Engineering from Chung-Ang University in 2019.



Hyung Woo Choi worked at the Department of Engineering, University of Cambridge, as a research associate from 2016 to 2021 as both a research associate and a senior research associate. He received a PhD in the Department of Materials Science and Engineering at Arizona State University in 2012.



Sung-Min Jung is currently working at the Department of Engineering, University of Cambridge, as a senior research associate. He was a research associate at the University of Cambridge from 2018 to 2020. Before moving to the University of Cambridge, he was a chief research engineer and a senior research engineer at LG Display R&D Centre, Korea, from 2004 to 2018. He received his PhD from Inha University in 2004.



Dong-Wook Shin received his PhD degree in 2014 from Sungkyunkwan University. He is currently working at the Department of Engineering, University of Cambridge, as a senior research associate. Before joining the University of Cambridge, he worked at the University of Exeter as a research associate.



Sanghyo Lee is currently working at the Department of Engineering, the University of Cambridge, since 2019 as a senior research associate. Previously, he worked at the University of Oxford from 2015 to 2019 as a research associate. He received his

PhD from the Department of Physics, Hanyang University, in 2015.



Jong Min Kim is currently Professor of Electrical Engineering of the Department of Engineering at the University of Cambridge. He was Chair of Electrical Engineering at the University of Oxford from 2012 to 2015. Before moving to academia, he was formerly Senior Vice President and Vice President in Samsung Electronics Corporate R&D Centre, Korea, for 13 years. He had previously held a variety of senior technology positions at the Samsung Group including Display, Materials, Energy and Electronics research/developments. Especially, he was the leader in developing and demonstrating a “world-first” QD display with AM matrix backplane that was published at Nature Photonics in 2009.

SUPPORTING INFORMATION

Additional supporting information may be found in the online version of the article at the publisher's website.

How to cite this article: Zhan S, Suh Y-H, Fan X-B, Yang J, Ni L, Kim Y, et al. Inkjet-printed multi-color arrays based on eco-friendly quantum dot light emitting diodes with tailored hole transport layer. *J Soc Inf Display*. 2022. 1–10. <https://doi.org/10.1002/jsid.1133>

APPENDIX A: EXPERIMENTAL METHODS

A.1 | Jetting waveform control of nozzle for printing

During printing, the jetting waveform of the nozzle had five stages with a maximum voltage of 15 V, as shown in Figure S1. The waveform was set into five stages to reset the piezo to a neutral position (Phase 0), draw ink from the cartridge (Phase 1), eject drops (Phase 2), and decrease voltage gradually (Phases 3 and 4), respectively.

A.2 | Device fabrication

The ink formulations including InP green- and red-based and ZnSeS blue-based QDs were prepared in cyclohexylbenzene. All the QDs used for the ink were synthesized. The fabrication procedures of the devices are

as follows. A pixelated ITO glass substrate (from Ossila ltd) with a sheet resistance of ~ 20 ohm/sq was cleaned with water, acetone, 2-propanol, each for 10 min. The ITO glasses were treated with UV-Ozone for 10 min to further clean the surfaces and enhance the wettability of PEDOT:PSS on the surfaces of the substrates. Then, PEDOT:PSS as the HIL was spin-coated and baked at 150°C for 20 min in the air. The PEDOT:PSS coated substrates were transferred to a nitrogen-filled glovebox for the following processes. Then TFB, PVK, t-TFB, t-PVK, TFB/PVK, or t-TFB/PVK was deposited at 3,000 rpm using solution with 8 mg/ml concentration in chlorobenzene and then baked at 150°C or 200°C for 30 min. For t-TFB and t-PVK, after the HTL film was coated, a drop of chlorobenzene was casted onto the film during spinning (3,000 rpm) before annealing. For TFB/PVK and t-TFB/PVK, PVK was spin-casted on the TFB or t-TFB film at 3,000 rpm and then annealed at 200°C for 30 min.

To deposit a QD layer by inkjet printing, the coated substrate was taken out of the glove box. Red, green (InP/ZnSeS), or blue (ZnTeSe/ZnSe/ZnS) QD ink was prepared in a mixed solvent (CHB/octane, 2% octane) with a concentration of 20, 12, or 7.5 mg/ml. The ink was printed based on the jetting waveform mentioned in Printing Waveform Control on substrates at 25°C or 50°C in the ambient condition. Afterward, samples were vacuum dried for 10 min and then annealed in a glovebox at

100°C for 10 min. Then ZnMgO (5% Mg, bought from Mesolight) of 25 mg/ml in ethanol was spin-coated and annealed at 120°C for 30 min. Finally, samples were sent into a thermal evaporation system where 100 nm Al was deposited with a rate of 0.15 nm/s and a vacuum level of 6×10^{-6} mbar. After fabrication, cover glasses and epoxy were used to encapsulate the printed Cd-free QLEDs.

A.3 | Bank fabrication

AZ5214 photoresist was spin-coated on ITO glasses at 4,000 rpm (height ~ 1.3 μm) and baked at 105°C for 1 min; then, samples were exposed with broadband ultraviolet light with a power of 10 mW/cm^2 for 25 s under a photomask; finally, samples were developed in MIF726 for 23 s, washed in DI water, and then dried by N_2 .

A.4 | Device characterization

To investigate the inkjet-printed QLED, EQE, current density/voltage (J-V), luminance/voltage (L-V), and EL spectra were measured by a photonic multichannel analyzer PMA-12 (Hamamatsu Photonics K.K.) connected with a Keithley 2400 source meter as the voltage and current source unit. PMA-12 is a compact spectral characterization instrument with high spectra sensitivity. By equipping an integrating sphere unit, the luminous efficiency of LEDs could be obtained accurately.