

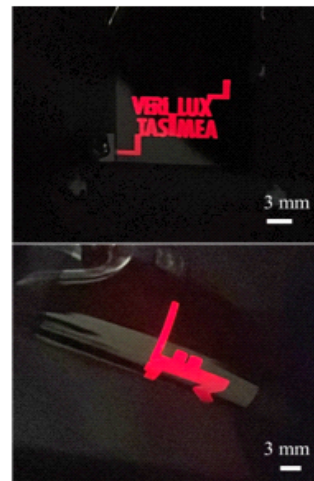
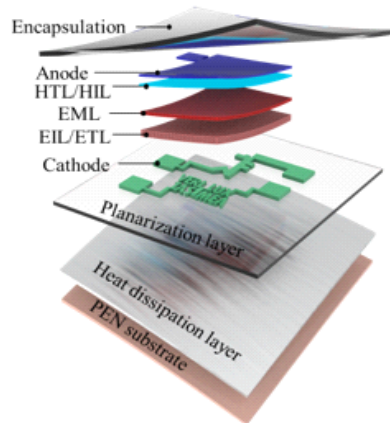
## RESEARCH

# Highly Bright and Efficient Flexible Quantum Dot Light-Emitting Diodes on Metal-Coated PEN Substrate

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## ABSTRACT

Quantum-dot (QD) based light-emitting diodes (QLEDs) have attracted significant attention in state-of-the-art wearable displays owing to the high form factor of QDs with excellent optical/electrical properties. These advantageous properties have led to notable advancements in QLEDs technology, making them flexible, lightweight, and compatible with human skin, which meets the requirements of wearable applications. To make the QLEDs wearable, however, alleviating the heat generated under high current is an urgent challenge. In this study, we present a flexible and bright top-emissive QLED fabricated on a plastic substrate with a thin (200 nm) heat dissipation layer. To preserve QLED performance even under sweaty conditions, we employed double layers of Parylene C/SU-8 for substrate planarization and device encapsulation. Furthermore, compared with typical wearable substrates that can hardly dissipate heat, the Al-metal induced flexible substrate can effectively dissipate the generated heat even under high current densities ( $2.3 \text{ A/cm}^2$ ), exhibiting a high luminance ( $\sim 366,000 \text{ cd/m}^2$ ) and a current efficiency ( $43.3 \text{ cd/A}$ ), respectively. The delicate device design of flexible QLEDs is anticipated to contribute to the advancement of next-generation wearable optoelectronics.

Key Words: Quantum dots, Quantum dot light-emitting diodes, Flexible, Wearable, Heat dissipation

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## 1. INTRODUCTION

Colloidal quantum-dot (QD) light-emitting diodes (QLEDs) have been considered as one of the promising candidates due to their excellent optoelectronic properties, such as size-dependent tunable bandgap, wide absorption, and high color purity with near-unity photoluminescence quantum yield (PL QY) [1-4]. In particular, the demand for flexible QLED displays as informative optoelectronic devices has significantly increased due to their high free-form factor, which is necessary in cutting-edge displays such as augmented/virtual reality, wearable glasses, and foldable/rollable smartphones that must be thin, light, flexible, and bendable [5-7]. This is because convenient and secure contact without the feeling of irritation is crucial for the comfort of the user in daily life. In this sense, portable lighting and display devices that can be user-interactive and freely available in the deformable shape is indispensable in human-lives [8-9].

There have been several advances in the development of wearable QLED displays that can be fabricated on flexible substrates. In 2015, Choi et al. reported the deformable and ultra-thin QLEDs display on curved and convoluted surfaces [10]. In 2020, Shin et al. reported waterproof and flexible InP based QLEDs on polyethylene naphthalate (PEN) substrate using CYTOP encapsulation layer [11]. In 2021, Kim et al. also reported 3-dimensional foldable QLEDs on a Parylene C that can be created using laser patterning and metal etch-stop layers with customized ablation thresholds [12]. Despite these achievements, however, most of the papers have focused on the fabrication method capable of making the devices thin, light, waterproof, and skin attachable. However, for QLEDs

that remain highly luminous and stable even under curved and deformable shapes, the low thermal conductivity ( $\kappa$ ) values ( $<0.3$  W/m·K) of conventional flexible substrates should be improved. Because of the low  $\kappa$  of the substrate, the light-emitting pixels in operation may experience thermal degradation at high current densities ( $J$ ) due to Joule heating. To resolve this challenge, several attempts have been made by the adoption of high- $\kappa$  substrates such as sapphire, Si, and Al-metal foil substrate, showing 46 W/m·K, 150 W/m·K, and 237 W/m·K, respectively [13-15]. However, these works have been attempted on inflexible substrates or on metal foils, so the stable operation of QLEDs is vulnerable to the repeatable deformable stress.

In this study, we have demonstrated bright and efficient QLEDs in an inverted top-emitting structure, which are fabricated on a PEN substrate with a thin (200 nm) Al-based heat dissipation layer. As a result of the enhanced heat dissipation property, the flexible top-emitting QLED (TQLED) exhibited a high luminance ( $L$ ) of  $\sim 366,000$  cd/m<sup>2</sup> and high current efficiency (CE) of 43.3 cd/A, while it stably operated at a high  $J$  up to 2,300 mA/cm<sup>2</sup>. Furthermore, we adopted Parylene C and SU-8 for both surface planarization and device encapsulation, making the flexible device waterproof. We posit that our approach to device design for developing flexible, wearable QLED displays provides valuable insights.

## 2. METHODS

### 2.1. Material Preparation

The CdSe/Cd<sub>1-x</sub>Zn<sub>x</sub>S QDs were synthesized in the same method reported previously [9]. To prepare the



ZnO nanoparticles, first, 80 mL of methanol and 2 g of zinc acetate dehydrate ( $\text{Zn}(\text{CH}_3\text{COO})_2 \cdot 2\text{H}_2\text{O}$ ) were mixed in a three neck flask. When the temperature grew up to  $60^\circ\text{C}$  in  $\text{N}_2$  atmosphere, potassium hydroxide dissolved in methanol ( $23.25 \text{ mg mL}^{-1}$ ) was added into the flask at the rate of  $238.5 \text{ mL h}^{-1}$ . After 150 min of reaction, the ZnO nanoparticles were sunk down for 12 h. After that, the solution was centrifuged at 4,000 rpm for 3 min to extract pure ZnO nanoparticles from the mixture solution. Finally, the ZnO nanoparticles were redispersed in 1-butanol.

## 2.2. Device Fabrication

To confirm the performance of the thin heat dissipation layer, we prepared the TQLED either on a typical PEN substrate or on the Al-coated PEN substrate. A 2.0- $\mu\text{m}$ -thick Parylene C layer was deposited using a Parylene deposition system (PDS 2010). After that, a 500-nm-thick epoxy (SU-8 2000.5, MicroChem) layer was spin-coated on the Parylene C layer for planarization for both substrates. On the substrates, a 100-nm-thick Ag layer was deposited as the reflective cathode electrodes by thermal evaporation. Then, the ZnO nanoparticles ( $20 \text{ mg/mL}$ ) were spin-coated at a rate of 2,000 rpm for 40 s, followed by annealing at  $100^\circ\text{C}$  for 30 min. The red-emitting CdSe/ $\text{Cd}_{1-x}\text{Zn}_x\text{S}$  QDs ( $20 \text{ mg/mL}$  in toluene) were deposited by spin-coating at a rate of 4,000 rpm for 30 s, and then dried at  $90^\circ\text{C}$  for 30 min. Then, 4,4'-bis(9-carbazolyl)-biphenyl (CBP, 50 nm),  $\text{MoO}_x$  (5 nm), dipyrzino[2,3-*f*:2',3'-*h*] quinoxaline-2,3,6,7,10,11-hexacarbonitrile (HAT-CN, 5 nm) and Ag (20 nm) were sequentially deposited by thermal evaporation as the hole transport layer, the hole injection layer, and the

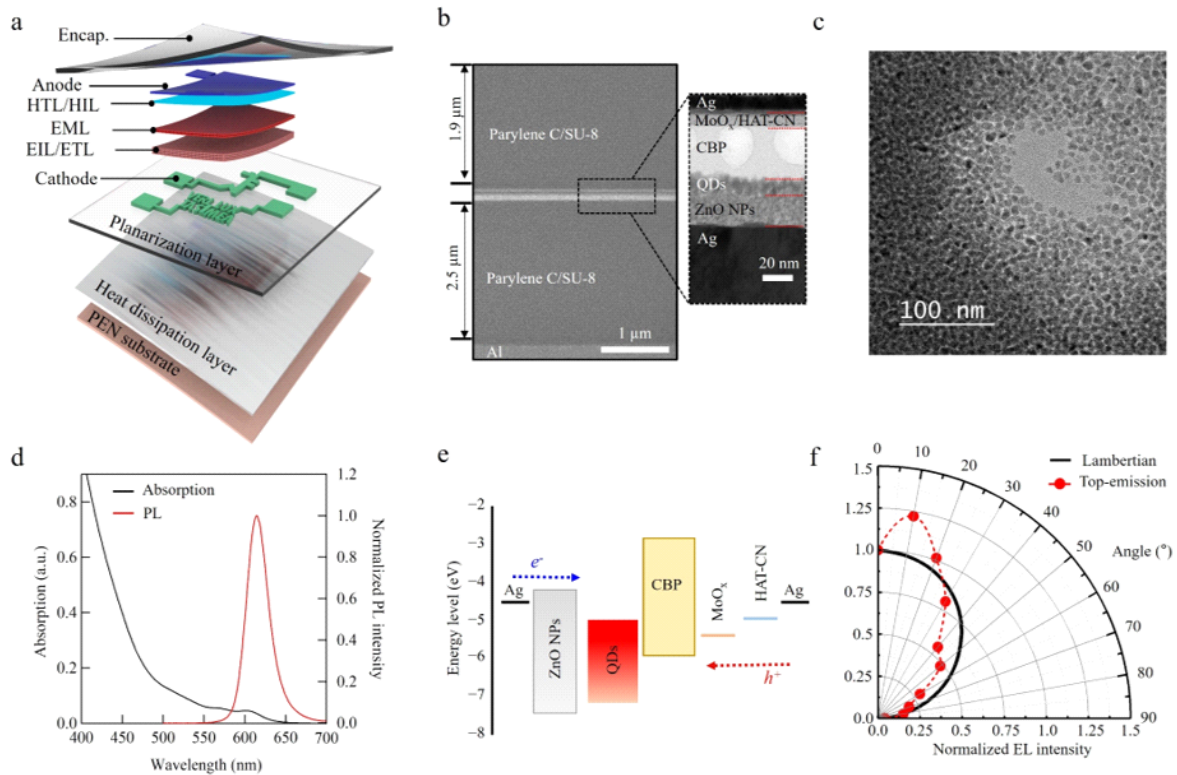
anode, respectively.

## 2.4. Characterization

The transmission electron microscopy (TEM) image was obtained using a Cs-corrected TEM (JEOL JEM-ARM200F). The surface morphology of the flexible substrates was obtained using an atomic force microscopy (AFM) (Park Systems NX-10). The  $J$ -voltage ( $V$ )- $L$  characteristics of the TQLEDs were measured using a Keithley 236 source-measure unit, a Keithley 2,000 multimeter, and a calibrated Si photodiode (Hamamatsu, S5227-1010BQ). The electroluminescence (EL) spectra were measured using a spectroradiometer (Konica-Minolta CS-2000). For the thermal simulations, a COMSOL Multiphysics software was used.

## 3. RESULTS AND DISCUSSION

Fig. 1(a) shows an exploded view of the flexible TQLED while Fig. 1(b) presents the cross-sectional TEM images of the TQLED device. We deposited a thin (200 nm) Al layer on the flexible PEN substrate as a heat dissipation layer. To alleviate the surface roughness of the substrate, Parylene C and SU-8 layers were deposited, because of their low temperature processability and high flexibility, in comparison to inorganic planarization/passivation layers, such as  $\text{SiO}_2$ ,  $\text{Si}_3\text{N}_4$ , and  $\text{HfO}_x$  [16-17]. For the TQLEDs, we utilized red CdSe/ $\text{Cd}_{1-x}\text{Zn}_x\text{S}$  QDs as the emissive layer (EML), of which an average diameter is  $13.2 \pm 1.2 \text{ nm}$ , as shown in Fig. 1(c). Fig. 1(d) shows their PL and absorption spectra; a PL peak wavelength at 614 nm, a full-width at half-maximum of 31 nm, and a PL QY of 80%. As shown in Fig. 1(b) and 1(e), the TQLED

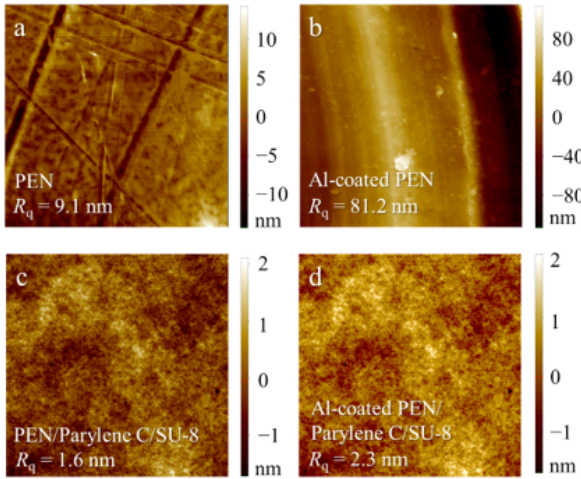


**Fig. 1.** Schematic device structure of the TQLED fabricated on a heat dissipation layer-coated PEN substrate. (a) Schematic view and (b) cross-sectional TEM image of the complete device (left) and the enlarged image of the TQLED (right). (c) Surface TEM image of the red-emitting CdSe/Cd<sub>1-x</sub>ZnxS QDs. (d) Absorption and PL spectra of the QDs. (e) Energy level diagram for the TQLED. and (f) The angle-dependent normalized EL intensity of the TQLEDs (red), along with the Lambertian distribution (black).

consist of Ag (85 nm)/ZnO nanoparticles (40 nm)/QDs (20 nm)/CBP (50 nm)/MoO<sub>x</sub> (5 nm)/HAT-CN (5 nm)/Ag (20 nm). For the efficient hole injection, we adopted MoO<sub>x</sub>/HAT-CN as a hole injection layer, as reported previously [14]. On the top of the device, the Parylene C/SU-8 bilayer was formed again for the water-resistant and ultra-thin encapsulation. Furthermore, to record the angular distribution of TQLEDs, we measured EL intensity along with the light-emission angle degrees from 0° to 90°. Fig. 1(f) shows that the TQLED emits light more to the normal direction deviating from the Lambertian profile, which is attributed

to the microcavity effect between two reflective metal electrodes [18]. For comparison, we also prepared the same device on a normal PEN substrate, and analyzed their surface roughness, heat dissipation properties, and TQLED device performance.

For bright and efficient flexible QLEDs, uniform surface morphology of flexible substrates is essential [19]. As shown in the AFM images in Fig. 2(a) and 2(b), the root-mean-square surface roughness ( $R_q$ ) values of the PEN and the Al-coated PEN substrates were measured to be 9.1 nm and 81.2 nm, respectively. Also, the peak-to-valley values were high as several

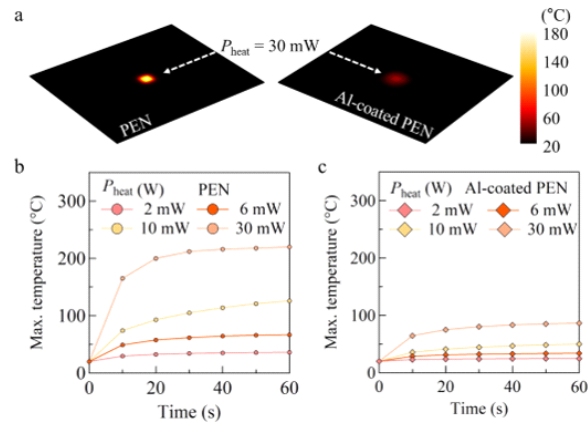


**Fig. 2.** Substrate morphologies of (a) PEN, (b) Al-coated PEN, (c) PEN/Parylene C, and (d) Al-coated PEN/Parylene C/SU-8, measured by AFM.

tens of nanometers for both cases. For the surface planarization of the substrates, a bilayer consisting of Parylene C/SU-8 was adopted, leading to uniform surface roughness with the  $R_q$  values of  $\sim 2$  nm for the two substrates, as shown in Fig. 2(c) and 2(d).

Next, we characterized the heat dissipation properties of the substrates. It is known that general polymeric materials have low  $\kappa$  values; for instance,  $\kappa$  of PEN, Parylene C, and SU-8 are  $\sim 0.19$  W/m·K, 0.08 W/m·K, and 0.2 W/m·K, respectively [20-22]. Thus, if Joule heat is generated during device operation, it impedes heat dissipation and can cause device degradation. However, by inserting a thin Al-based heat dissipation layer into the substrate, Joule heat can be effectively dissipated to prevent device degradation owing to the high  $\kappa$  of Al (237 W/m·K). In order to clarify the influence of the thin Al layer, placed between the PEN substrate and the Parylene C/SU-8 planarization layer, on the heat dissipation, we conducted

thermal simulations using a COMSOL Multiphysics software. In this simulation, heat radiation and air-cooling were ignored. Fig. 3(a) shows the simulated temperature profile of the pixels on the respective substrate, where the pixel was considered as a Joule heat source with the applied power ( $P_{\text{heat}}$ ) of 30 mW. For the accuracy of  $P_{\text{heat}}$ , the optical output power should be subtracted from the applied electrical power (i.e., current  $\times$  voltage). However, since the optical power is negligible when compared to the electrical power, we simply assumed that  $P_{\text{heat}}$  is identical to electrical power [23]. The pixel with a dimension of  $1.4 \text{ mm} \times 1.4 \text{ mm}$  were positioned at the center of the substrate with an area of  $20 \text{ mm} \times 24 \text{ mm}$ , which are identical to the devices. The simulation results of the maximum pixel temperature with varying  $P_{\text{heat}}$  on the PEN and Al-coated PEN substrates are plotted in Fig. 3(b) and 3(c), respectively. It is obvious that the temperature on the Al-coated PEN substrate is much lower than that



**Fig. 3.** Thermal simulations of heat dissipation in the TQLEDs with two substrates. (a) Temperature profile of the QLED pixels when  $P_{\text{heat}}=30$  mW. The maximum temperature at various  $P_{\text{heat}}$  over time in (b) the PEN and (c) the Al-coated PEN substrates.

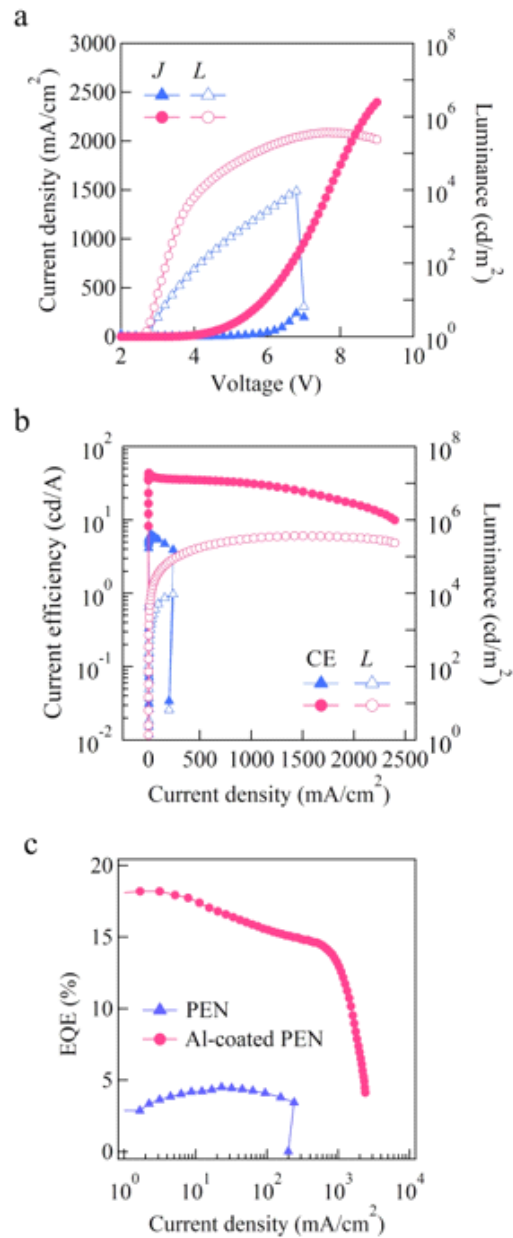


in the normal PEN substrate. For example, when  $P_{\text{heat}}$  is 30 mW, the pixel temperature on the PEN substrate was increased over 200°C which is high temperature sufficient to deteriorate the TQLED [13,24-25]. Meanwhile, the maximum temperature of the pixel on the heat dissipation layer-coated PEN substrate reached below 100°C at the same condition, verifying the heat dissipation ability of the thin Al layer. Therefore, it is expected that the TQLED fabricated on the Al-coated substrate can endure much higher current densities than that prepared on the normal PEN substrate.

To confirm this, we measured the device performance of the TQLEDs fabricated on the PEN and Al-coated PEN substrates. As shown in Fig. 4(a), the TQLED on a normal PEN substrate presents catastrophic failure at  $J$  of  $\sim 240 \text{ mA/cm}^2$  at 6.8 V, which corresponds to  $\sim 30 \text{ mW}$  of  $P_{\text{heat}}$ . Since the simulation showed that 30 mW of  $P_{\text{heat}}$  can result in a pixel temperature  $>200^\circ\text{C}$ , the failure is attributed to the Joule heat. On the other hand, the TQLED fabricated on the substrate with the heat dissipation layer could endure much higher  $J$  of up to 2,300  $\text{mA/cm}^2$  (9 V), which generates  $P_{\text{heat}}$  of  $>400 \text{ mW}$ . Furthermore, CE, the maximum  $L$  ( $L_{\text{max}}$ ), and external quantum efficiency

**Table 1.** Performance of flexible QLEDs.

| Substrate                 | EL peak (nm) | $L_{\text{max}}$ ( $\text{cd/m}^2$ ) | CE (cd/A) |
|---------------------------|--------------|--------------------------------------|-----------|
| Parylene C/SU-8 [26]      | $\sim 540$   | 44,700                               | 4.5       |
| Parylene C/SU-8 [12]      | $\sim 550$   | 50,500                               | -         |
| PET [27]                  | 606          | 106,890                              | 16.3      |
| Al foil [5]               | 624          | 39,880                               | 3.6       |
| PEN (this work)           |              | 9,420                                | 6.2       |
| Al-coated PEN (this work) | 621          | 366,000                              | 43.3      |



**Fig. 4.** Device performance of the TQLEDs in terms of (a) the  $J$ - $V$ - $L$ , (b) the  $\text{CE}$ - $J$ - $L$ , and (c)  $\text{EQE}$ - $J$  characteristics on the PEN (blue triangle) and Al-coated PEN (pink circle) substrates.

(EQE) were significantly improved from 6.2 cd/A, 9,420  $\text{cd/m}^2$ , and 4.51% to 43.3 cd/A, 366,000  $\text{cd/m}^2$ , and 18.19%, respectively, as shown in Fig. 4(b). These

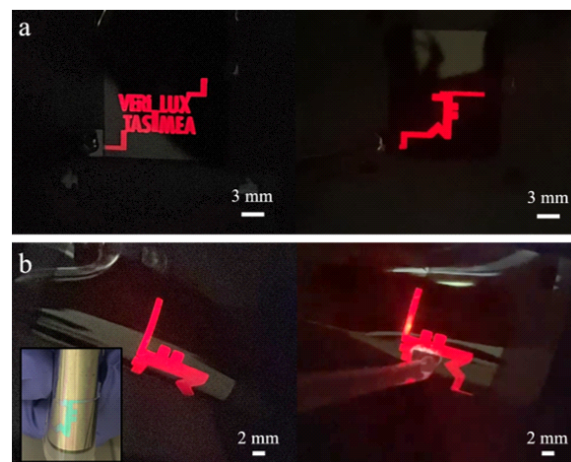


results clearly show that the insertion of the thin metal-based heat dissipation layer is an excellent strategy to improve the performance of flexible QLEDs based on any types of plastic substrates, such as poly(ethylene-terephthalate) (PET), polyethersulfone, and polyimide. The device performance of a few flexible QLEDs are compared in Table 1.

In addition to the improved device performance of the TQLEDs, we demonstrate the waterproof property which is indispensable for wearable application. We fabricated the devices with larger emission patterns on the substrates with a dimension of 30 mm × 40 mm. As displayed in Fig. 5, the letters and symbol patterns on the device stably emit red light while curled up in the pen. Additionally, owing to the Parylene C/SU-8 passivation layer, the device shines well even with a water droplet. Based on these results, we believe that our strategies for bright and efficient flexible QLEDs would be helpful for the development of human interactive display in near future.

#### 4. CONCLUSION

In conclusion, we have demonstrated highly bright, efficient, and water-resistive flexible TQLEDs that hold promise for a range of applications. To address the low- $\kappa$  of typical plastic-based flexible substrates, we developed a PEN substrate with a thin metal heat dissipation layer to improve the heat dissipation property. As a result, our TQLEDs exhibited significantly higher brightness and current efficiency compared to previously reported flexible QLEDs. Furthermore, we have successfully showed the large-area devices that can operate even in wet conditions, meeting the necessary requirements for flexible and



**Fig. 5.** Demonstration of the large-area TQLED display fabricated on Al-coated PEN substrates, showing the (a) “VERITAS LUX MEA” (left) and the university symbol (right), and its operation under (b) bent (left) and with a water droplet (right). The inset shows the device rolled on a pen without electrical bias.

wearable displays. We believe that our methodology will provide helpful insight and contribute to the development of wearable optoelectronic devices.

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##### Author Contributions

G.W.B. and H.S. contributed equally to this work. G.W.B., H.S., and J.K. proposed idea and designed the experiments. G.W.B. and H.S. fabricated and characterized the TQLED devices. D.H. and W.K.B. designed and synthesized QDs. T.L. assisted the demonstration of large-area devices. G.W.B. and H.S. prepared the manuscript and J.K. revised it. All authors read and approved the final manuscript.

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#### Declarations of Competing Interests

The authors declare that they have no competing interests.

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