

Electronic Supplementary Information

## Influence of Spinodal Decomposition-based Phase Separation in Hybrid Polymer Hole Transport Layer on Electroluminescent Quantum Dot Light-Emitting Diodes

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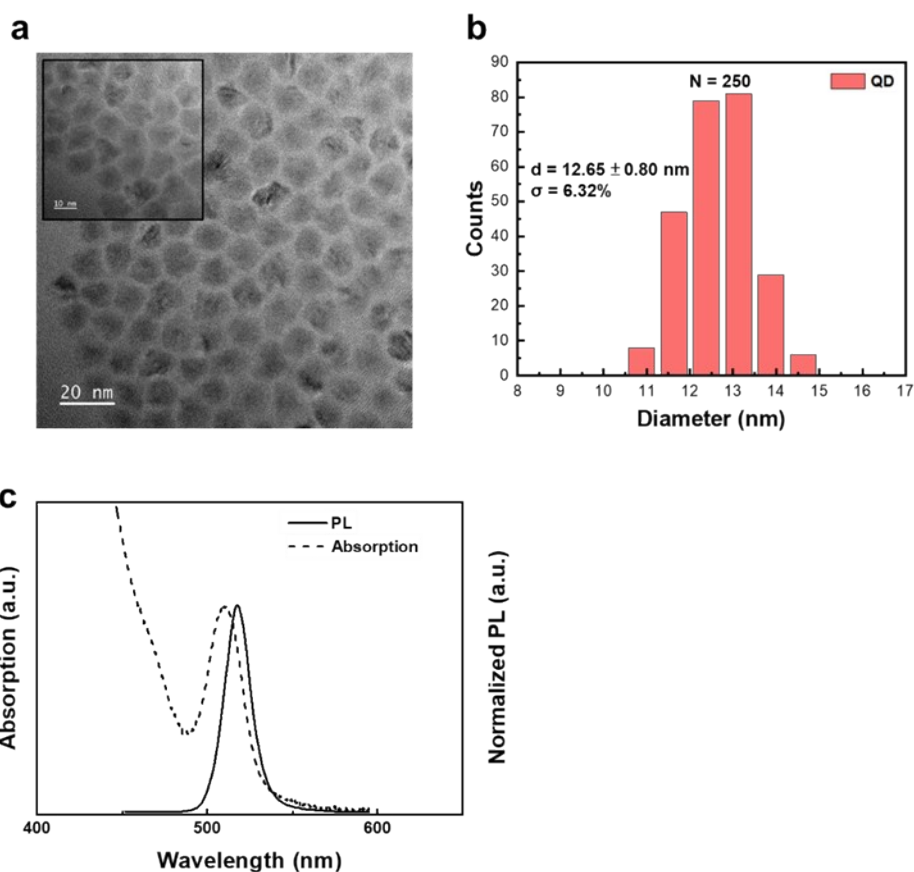
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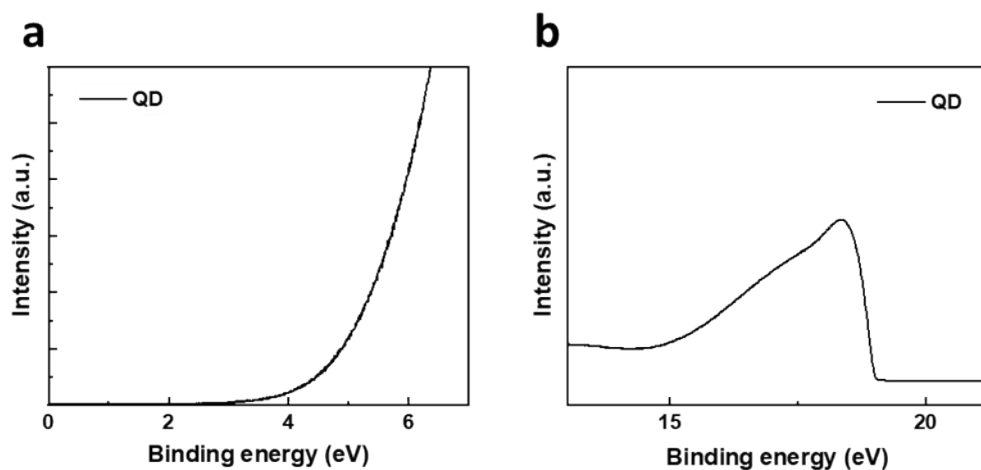
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**Fig. S1** (a) TEM image, (b) size distribution and Optical absorption and PL spectra of as-obtained green QDs. The non-spherical QDs are alloy ZnCdSeS with protrudes passivating the crystal facets predominately at (111) and (220).



**Fig. S2** UPS spectra of green QDs. (a) Onset region in valence-band region and (b) the secondary cut off region.

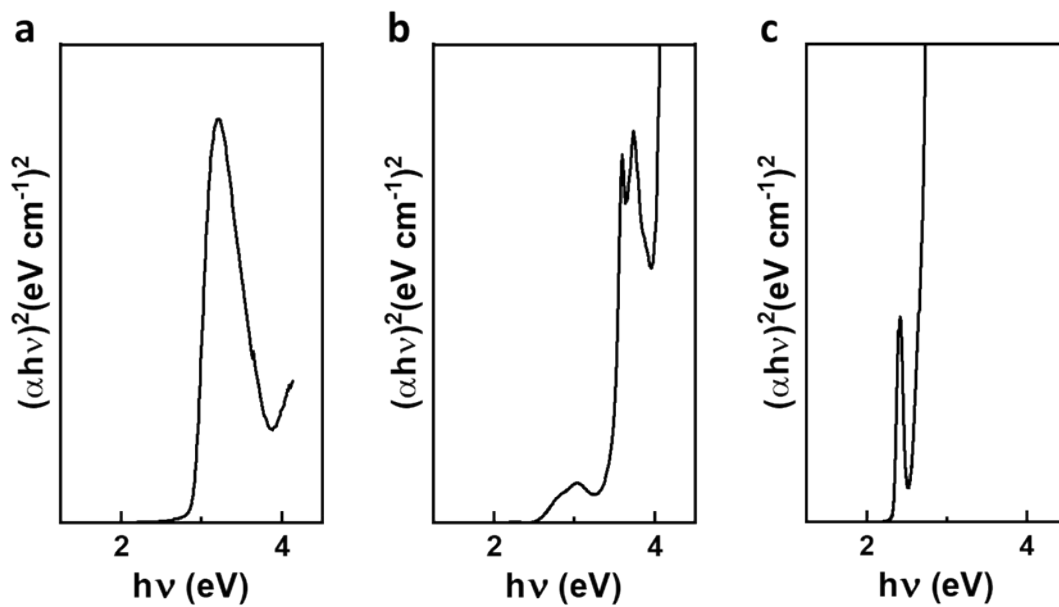


Fig. S3 Tauc plot of (a) TFB (b) PVK and (c) green QD.

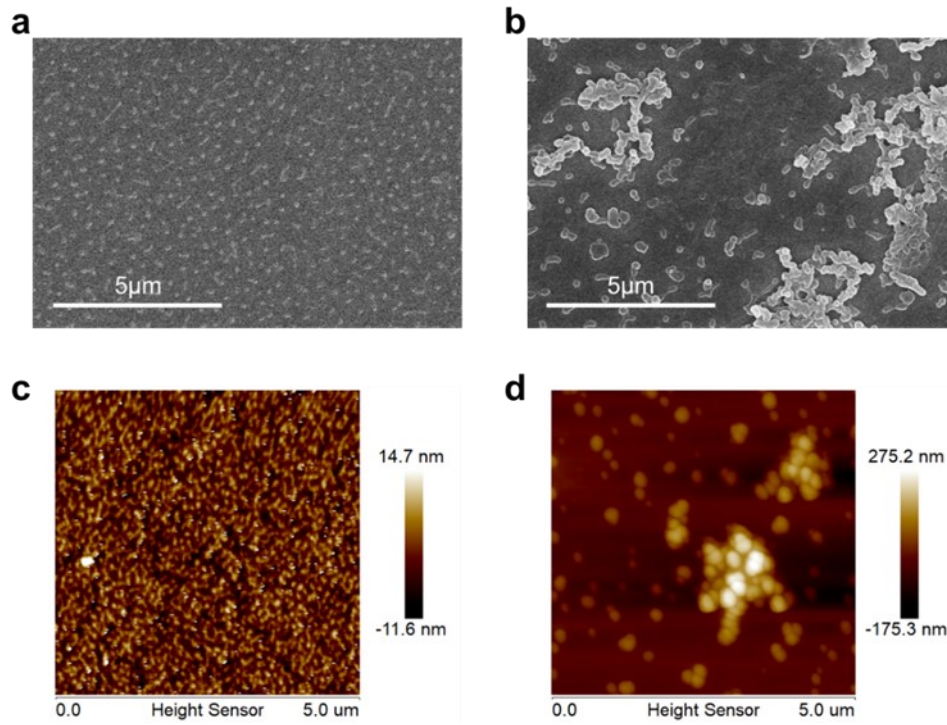


Fig. S4 SEM image of (a) spin-coating 1,4-dioxane on TFB film, (b) spin-coating PVK solution with 1,4-dioxane as solvent on TFB film and cause erosion. AFM image of (c) spin-coating 1,4-dioxane on TFB film, (d) spin-coating PVK solution on TFB and form non-uniform film.

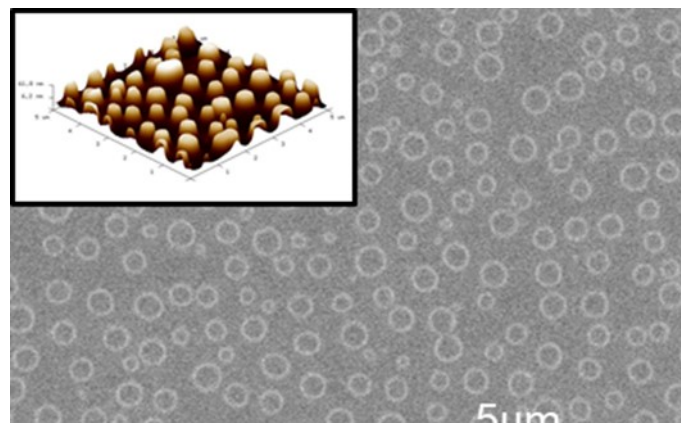
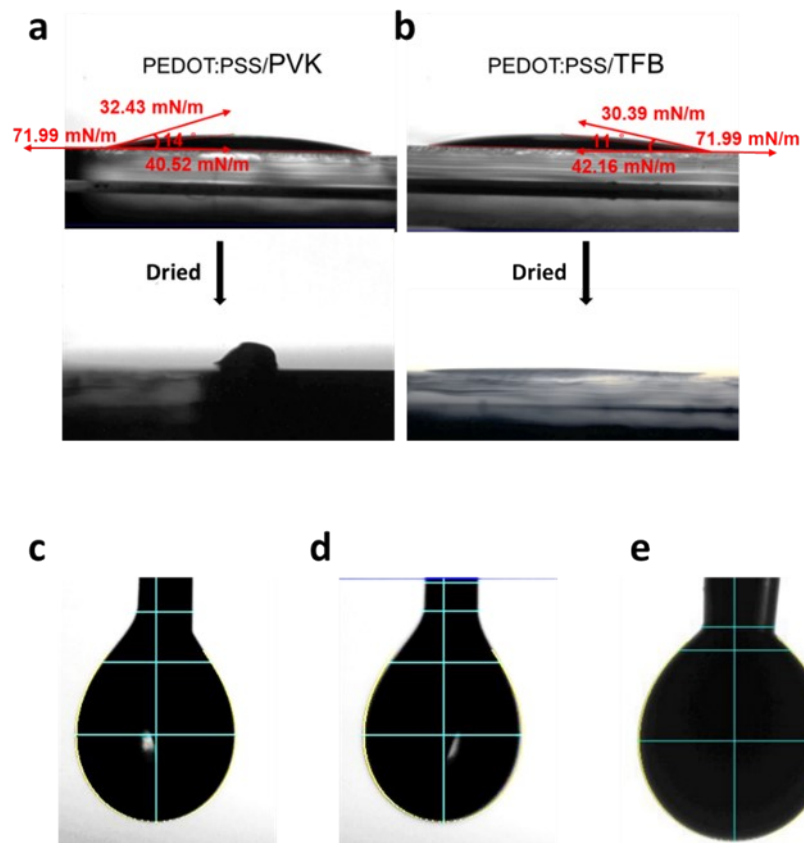


Fig. S5 SEM and AFM images of TFB-50 vol% films on the ITO/PEDOT:PSS substrates.



**Fig. S6** Contact angle and surface tension of (a) PVK solution and (b) TFB solution on PEDOT:PSS film. Pendant drop method of (c) PVK droplet, (d) TFB droplet and (e) PEDOT:PSS droplet.

**Table S1.** Surface tension of PEDOT:PSS film, TFB droplet and PVK droplet.

	$\cos\theta$	Surface tension (mN/m)
PEDOT:PSS	1	71.99
TFB-25 vol %	0.927	33.39
TFB-75 vol %	0.883	32.43

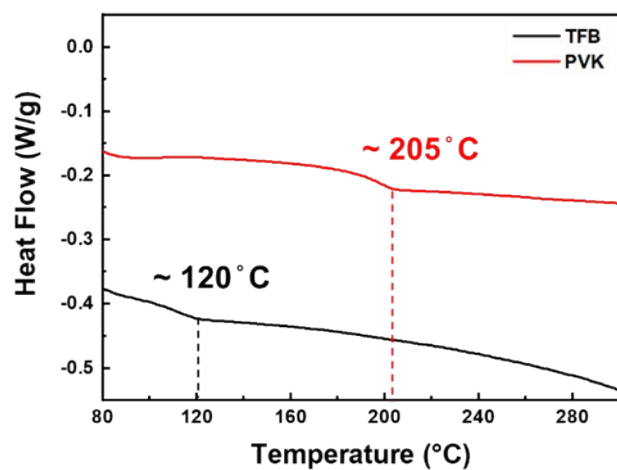


Fig. S7 DSC analysis of PVK and TFB.

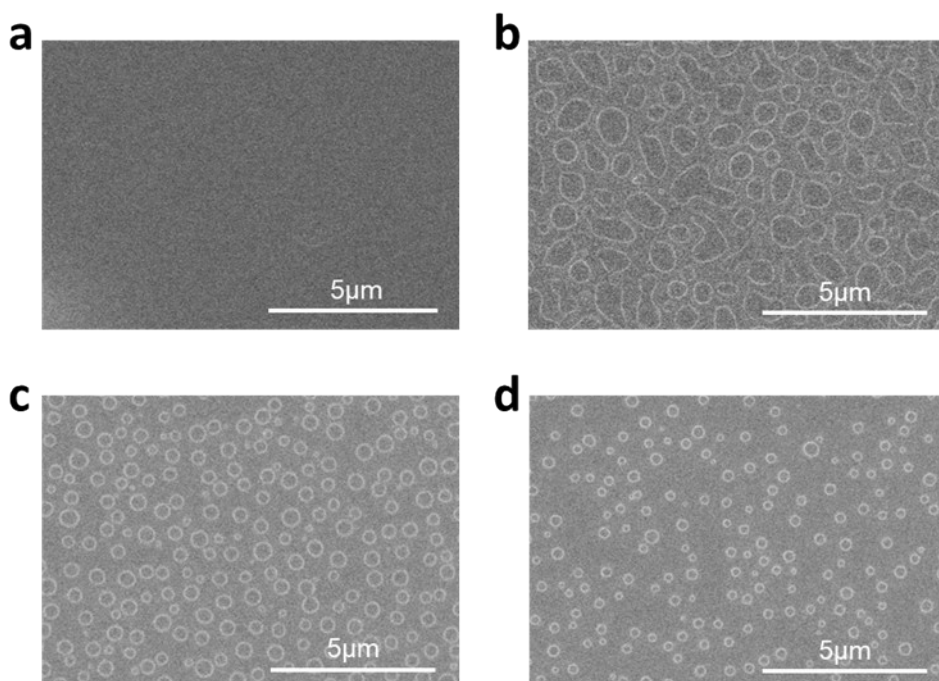


Fig. S8 SEM images of (a) TFB-10 vol%, (b) TFB-25 vol%, (c) TFB-50 vol%, (d) TFB-75 vol% films on the ITO/PEDOT:PSS substrates.

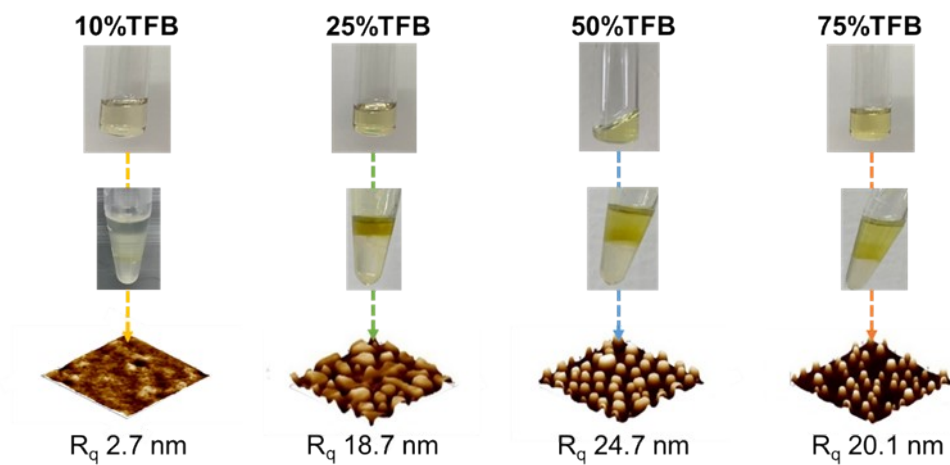


Fig. S9 The path of homogeneous solution to phase separation film in PVK-TFB-Toluene ternary phase diagram.

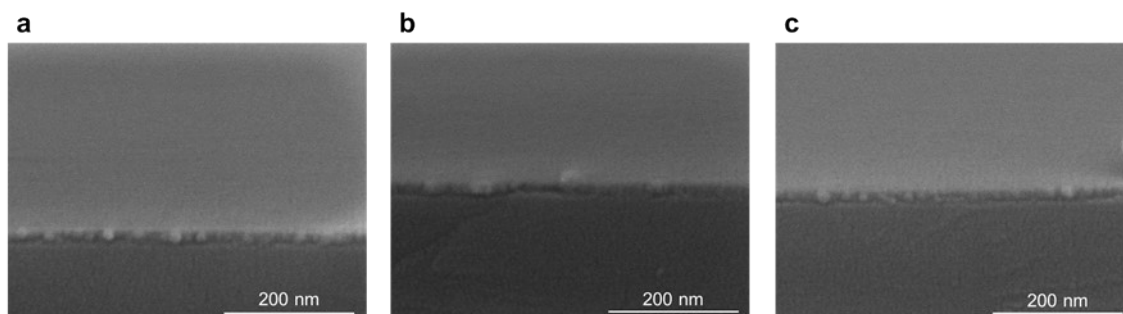


Fig. S10 Cross section SEM images of (a) PVK, (b) TFB and (c) 10% TFB- 90% PVK layers. The thickness of PVK, TFB, and TFB/PVK films are measured to be approximately 30, 27, 29 nm, respectively.

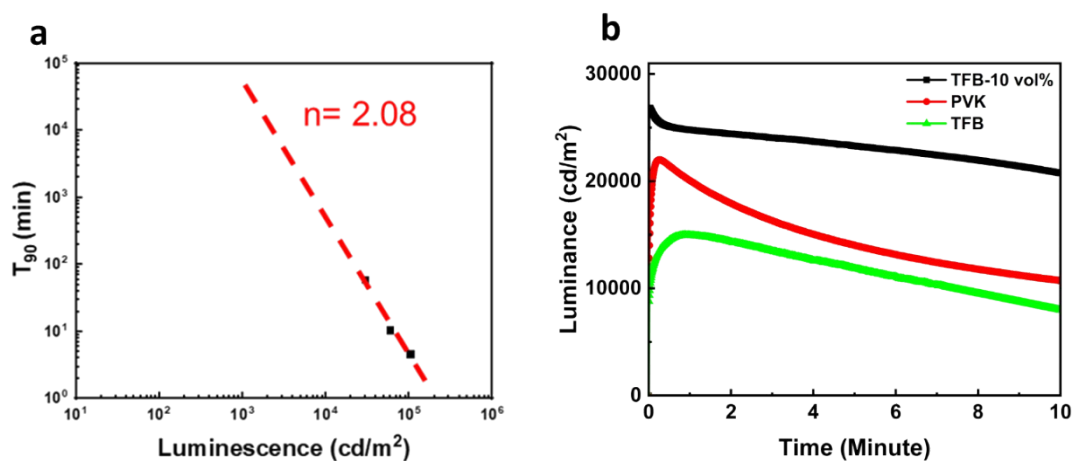


Fig. S11 (a) The acceleration lifetime of QLED (device with hybrid HTL of 10% TFB in PVK), equivalent to 100 nits for 129,374 h. (b) The lifetime of three devices as optimal hybrid HTL, pure PVK and TFB.

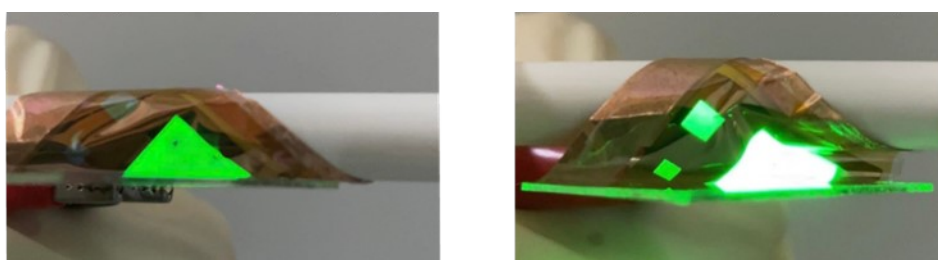
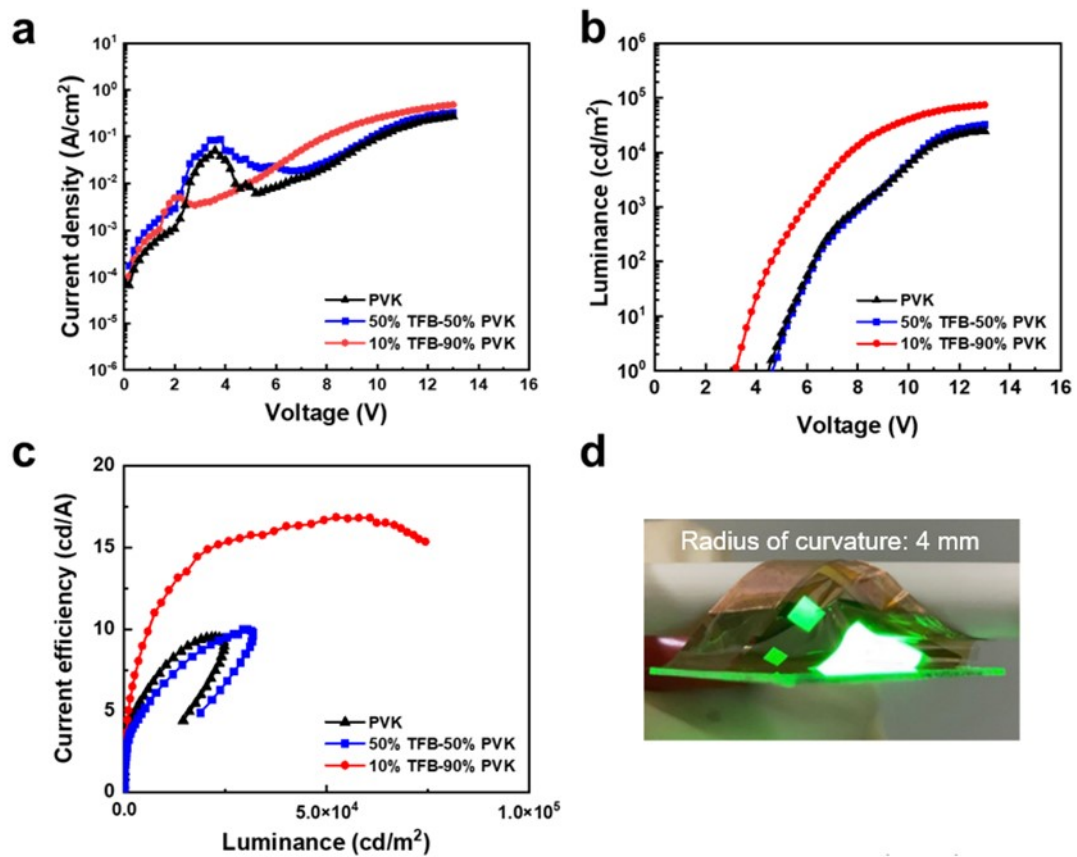


Fig. S12 TFB-PVK hybrid flexible QLED device with (a) and without (b) phase separation upon the bending. In the left photo, the dark spots show non-uniform regions, which are observed in the large area device. The small area EL devices have lower brightness and the non-uniform regions are hardly observed by naked eyes. In the right photo, none of non-uniform regions are observed in both large and small area film by either naked eyes or photos.



Fig. S13 PVK and Hybrid HTL flexible QLED devices. (a) Current density-voltage (J-V) characteristics. (b) Voltage-luminance characteristics. (c) Current efficiency-luminance characteristics. (d) Photo of the flexible devices.





**Table S2.** Comparison of the performance of atmosphere-fabricated QLEDs. (the device with green QDs and all layers fabricated under atmosphere conditions)

Device structure	HTL	Turn-on voltage (V)	Maximal luminance (cd/m <sup>2</sup> )	Maximal current efficiency (cd/A)	Process condition	Reference
PEDOT/HTL/QDs/ZnO	Poly TPD	6	3050	2.8	Atmospheric fabrication (All layers, except metal cathode)	Adv. Optical Mater. 2020, 8, 1901429
PEDOT/HTL/QDs/ZnO	PVK:TAPC	3.3	24,800	26.2	Atmospheric fabrication (All layers, except metal cathode)	RSC Adv., 2017, 7, 43366-43372
Tandem structure		6.1	115,500	121.5	Atmospheric fabrication (All layers, except Al, HATCN and MoO <sub>3</sub> )	ACS Nano 2018, 12, 1, 697–704
PEDOT/HTL/QDs/MgZnO	PVK:TFB	3.1	276,600	26.8	Atmospheric fabrication (All layers, except metal cathode)	Our work