Supporting information

Advanced HIL strategies in QLEDs:V₂O₅ and PEDOT:PSS dual-

layer for charge balance and electron leakage prevention

Han Bin Cho,^a Ha Jun Kim,^a Noolu Srinivasa Manikanta Viswanath,^a Tuhin Samanta,^a Jeong Wan Min,^a Sung Woo Jang,^a Yong Min Park,^a Se Hyuk Jang,^a Heesun Yang,^{*b} and Won Bin Im^{*a}

^aDivision of Materials Science and Engineering, Hanyang University, 222 Wangsimni-ro, Seongdong-gu, Seoul, 04763, Republic of Korea

^bDepartment of Materials Science and Engineering, Hongik University, 94 Wausan-ro, Mapogu, Seoul 04066, Republic of Korea

Corresponding Author E-mail: hyang@hongik.ac.kr, imwonbin@hanyang.ac.kr

Experimental section

Materials Vanadium (V) oxytriisopropoxide was purchased from Sigma-Aldrich. PEDOT:PSS (AI 4083) was purchased from Heraeus. Poly (9-vinyl carbazole) (MW = 90,000) was purchased from Acros. Isopropyl alcohol (99.5%) was purchased from Daejung Chemicals & Metals Co. Ltd. Aluminum (99.999%) was purchased from ITASCO. All chemicals were used without any additional purification steps.

Preparation of V₂O₅ HIL The precursor solution was prepared by diluting vanadium oxytriisopropoxide with isopropyl alcohol. To fabricate the V₂O₅ HIL, spin-coating was employed at 4,000 rpm for 45 s, followed by preheating at 150 °C for 5 min. The RTA procedures were conducted under ambient air conditions. The preheated V₂O₅ HIL sample was further subjected to heating using tungsten lamps for 30 s at heating rates ranging from 10 to 20 K s⁻¹ per second (**Fig. S1**).

Fabrication of QLED ITO substrates were sonicated in acetone and isopropyl alcohol for 30 min. It was then subjected to a cleaning process involving UV-ozone treatment for 15 min. Subsequently, a V_2O_5 HIL was deposited, as described above. PEDOT:PSS was spin-coated at 3,000 rpm for 45 s and annealed at 150 °C for 30 min. A solution of PVK (10 mg/mL in chlorobenzene) was spin-coated at 3,000 rpm for 45 s and annealed at 160 °C for 30 min. Subsequently, green CdSe/ZnSeS/ZnS QDs (optical density of 1.0, at 520 nm) were spin-coated at 3,000 rpm for 20 s and annealed at 70 °C for 30 min. The ZnMgO NPs (in ethanol at a concentration of 30–40 mg/ml) were spin-coated at 3,000 rpm for 60 s. Finally, the fabrication of the QLED was completed after thermal evaporation of a 100 nm-thick aluminum cathode.

Characterization X-ray diffraction (XRD) was used to assess the crystalline phases of the V_2O_5 films using a SmartLab instrument (Rigaku). TEM using a JEOL JEM 2100F instrument was used to determine the layer thickness and calculate the lattice distances. XPS was performed using a K-alpha instrument to determine the chemical composition of the V_2O_5 film. Surface analysis of V_2O_5 was performed using scanning electron microscopy (SEM) (Nova NanoSEM 450, FEI). Conductive atomic force microscopy (C-AFM) with an AFM instrument from Park Systems (NX20) was used to study the surface morphologies and conductive properties of V_2O_5 . Ultraviolet photoelectron spectroscopy (UPS) was used to evaluate the band levels of each layer using an XPS theta probe machine (Thermo Fisher Scientific Co.) equipped with a He¹photon source at 21.2 eV. UV–VIS spectra were acquired using a UV/VIS spectrophotometer (Optizen POP-S). The current density-voltage-luminance (J-V-L) characteristics of the devices were measured using a spectroradiometer (CS-2000, Konica Minolta) with a Keithley 2400 source meter under ambient conditions. Optical simulations were performed using commercial software (Setfos, Fluxim). The refractive index measurements were conducted via spectroscopic ellipsometry using an Elli-SE(UV)-FM8 instrument from Ellipso Technology Company, Ltd.



Fig. S1 Schematic diagram for synthesizing A4 and A8 thin films.



Fig. S2 XRD patterns of pre-heated sample.



Fig. S3 Line-scanning intensity profiles of V_2O_5 film.



Fig. S4 Raman spectra of V_2O_5 thin film.



Fig. S5 Conductive AFM and line profile of A8 low crystalline area.



Fig. S6 Dispersive spectroscopy (EDS) mapping images.



Fig. S7 Density of states of (a) V_2O_5 and (b) V_2O_5 with one electron.



Fig. S8 Urbach plots of A4 and A8.



Fig. S9 Device lifetime of PEDOT:PSS-based QLED and A8/PEDOT:PSS QLED, showing T50 under a constant current density at an initial luminance of 1000 cd/m².

| Materials | Transmittance [%] | Refractive index |
|-----------|----------------------|---------------------|
| Α4 | 98.74 | 1.86 |
| A8 | 98.46 | 1.90 |

Table S1Transmittance and refractive index of A4 and A8.