

# DERIVATIVES OF IMIDAZOLE AND CARBAZOLE AS BIFUNCTIONAL MATERIALS FOR ORGANIC LIGHT-EMITTING DIODES

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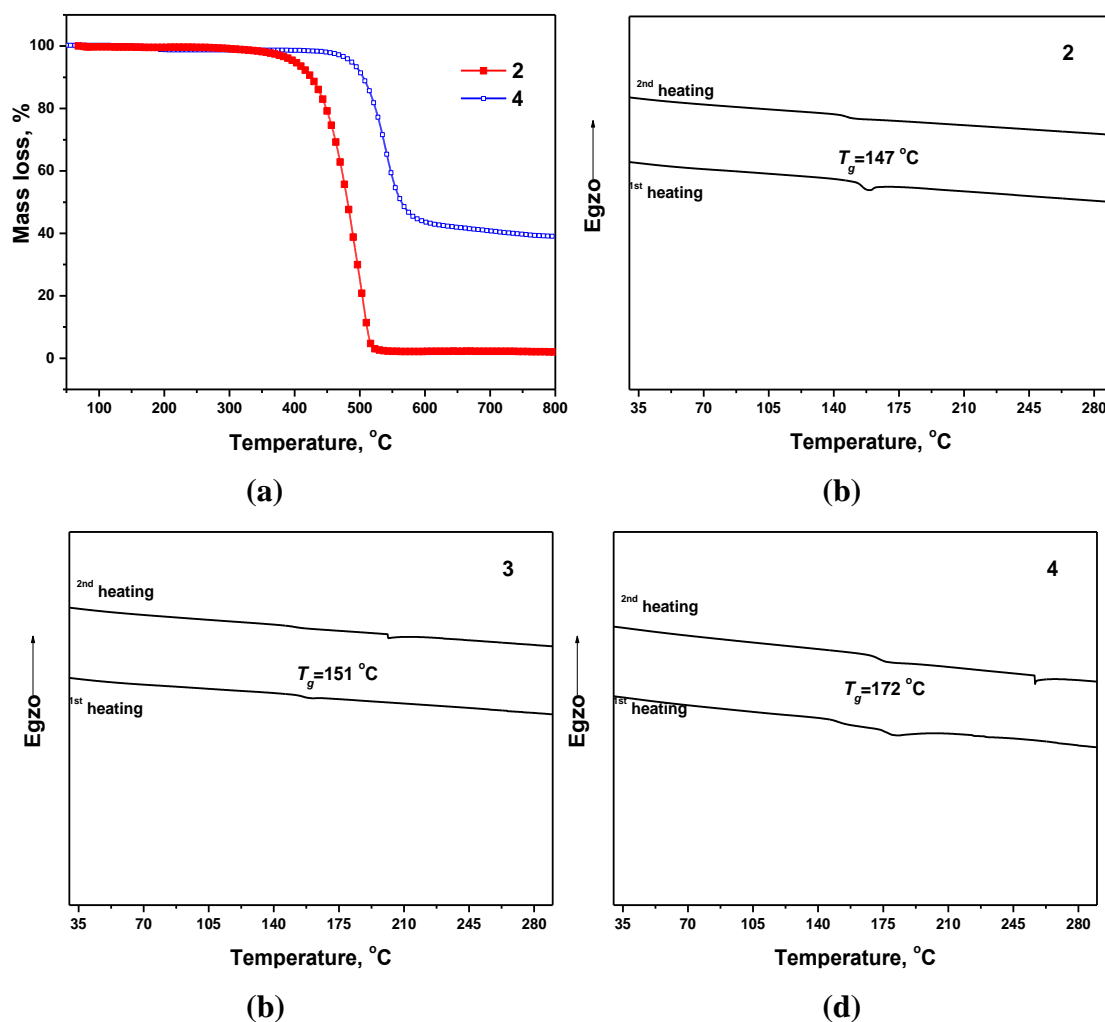
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## 1.1. Instrumentation

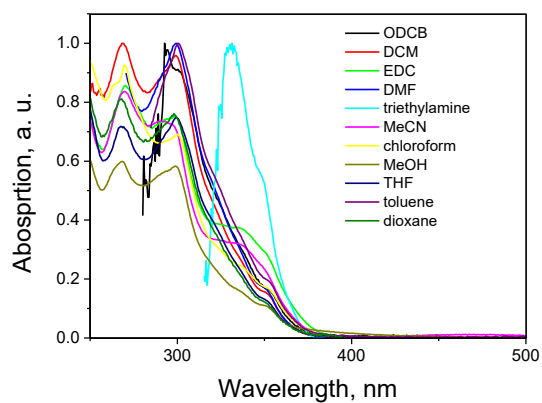
$^1\text{H}$  NMR and  $^{13}\text{C}$  NMR spectra were obtained of solutions in deuterated chloroform ( $\text{CDCl}_3$ - $d_6$ ) or dimethyl sulfoxide ( $\text{DMSO}-d_6$ ) with a Bruker Avance III spectrometer. The data are given as chemical shifts in  $\delta$  (ppm) and tetramethylsilane was used as an internal standard. IR spectra were recorded by a Vertex 70 Bruker spectrometer equipped with an ATR attachment with a diamond crystal over frequencies of  $600\text{--}3500\text{ cm}^{-1}$  with a resolution of  $5\text{ cm}^{-1}$  over 32 scans. Mass spectra were obtained on a Waters ZQ 2000 mass spectrometer. Melting point (mp) was determined using Electrothermal Mel-Temp melting point apparatus. Thermogravimetric analysis (TGA) was performed on a TA Instruments Q50 apparatus at a heating rate of  $20\text{ }^\circ\text{C}/\text{min}$  under nitrogen atmosphere. Differential scanning calorimetry (DSC) measurements were performed on a 100 TA Instruments Q2000 thermal analyzer at a heating/cooling rate of  $10\text{ }^\circ\text{C}/\text{min}$  under nitrogen atmosphere. Photophysical properties of  $10^{-4}$  M solutions in tetrahydrofuran and pure layers of the synthesized compounds were recorded using a Perkin Elmer Lambda 35 and an Edinburgh Instruments FLS980 spectrometers. Cyclic voltammetry (CV) measurements were carried out using an Autolab potentiostat PGSTAT20 in a three electrode cell using platinum rod as a counter electrode, glassy carbon as working

electrode and silver wire as the reference electrode. The experiments were carried out in dry dichloromethane solution containing 0.1 M tetrabutylammonium perchlorate as electrolyte at room temperature under nitrogen atmosphere at a scan rate 50 mV/s. The measurements were calibrated using the internal standard ferrocene/ferrocenium ( $\text{Fc}/\text{Fc}^+$ ). The ionization potentials ( $\text{IP}_{\text{PE}}$ ) of the films of the synthesized compounds were measured by UV photoelectron spectrometry in air[1]. Hole drift mobility of the materials was estimated by a time-of-flight (TOF) method. The samples were fabricated with structures of ITO/compound/Al. The thicknesses of the layers for TOF measurements were 2.6, 3.2, 2.7, 5.4  $\mu\text{m}$  for 1, 2, 3 and 4, respectively. Commercial indium tin oxide (ITO)-coated glass was used as a substrate, which was first cleaned chemically using a bath of distilled water and acetone. Organic and Al films were sequentially vacuum-deposited onto precleaned ITO-coated glass substrates under the vacuum of  $2\text{--}5 \cdot 10^{-6}$  mBar using vacuum equipment from Kurt J. Lesker in-built in an MB EcoVap4G glove box. In the time of flight (TOF) experiments, charges were generated by a pulsed third-harmonic Nd:YAG laser EKSPLA NL300 working at a pulse duration of 3–6 ns and the wavelength of 355 nm. Electric fields were applied by a Keithley 6517B electrometer. A digital storage oscilloscope Tektronix TDS 3032C was used to record TOF transients. The drift mobility was calculated by using the formula  $\mu = d^2/U \cdot t_t$ , where  $d$  is the layer thickness,  $U$  the surface potential at the moment of illumination, and  $t_t$  is the transit time which was taken from the TOF transients. Current–voltage curves were recorded using a Keithley 2400 Source Meter. A xenon lamp fitted with an AM1.5 global filter was used for illumination. Silicon photodiode was used to calibrate the light source to give 1 sun light intensity. Voltage was cyclically changed from 0 to 1 V and to -1 V under illumination or in the dark. A Keithley source meter 2400-C was utilized for recoding of the current density–voltage characteristics. The current density–luminance characteristics were estimated using a calibrated silicon photodiode with the 6517B Keithley electrometer. Electroluminescence (EL) spectra were

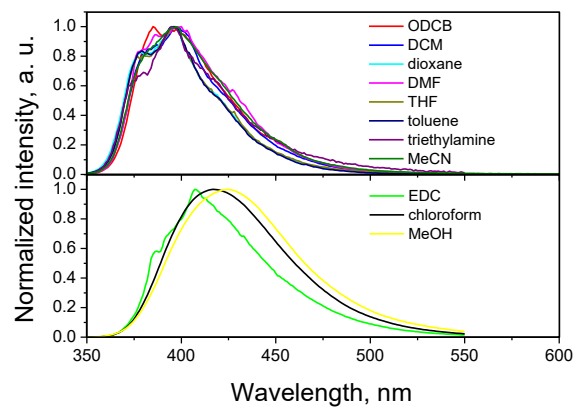
recorded by an Avantes AvaSpec-2048XL spectrometer. The current, power and external quantum efficiencies were estimated utilizing the current density, luminance, and EL spectra as reported earlier [2].



**Figure S1.** (a) TGA curves of compounds **2** and **4**; (b-d) DSC curves of compounds **2–4**

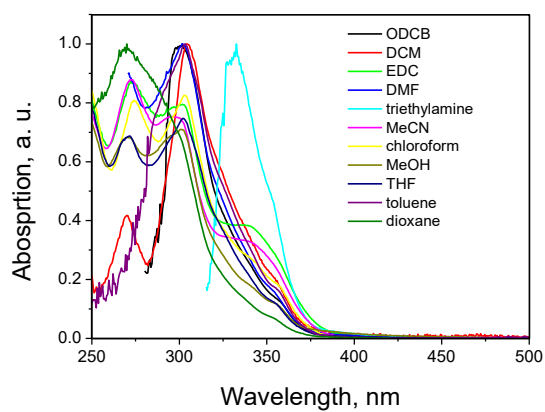


(a)

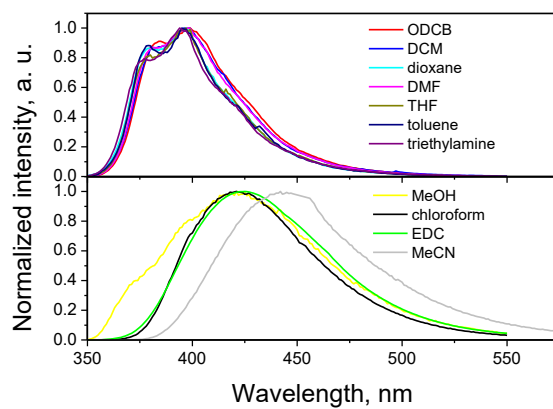


(b)

**Figure S2.** Absorption (a) and PL (b) spectra of **1** solutions

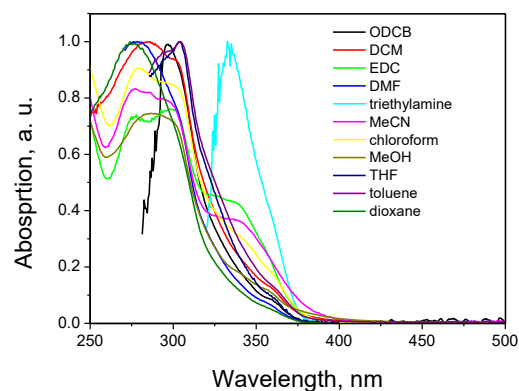


(a)

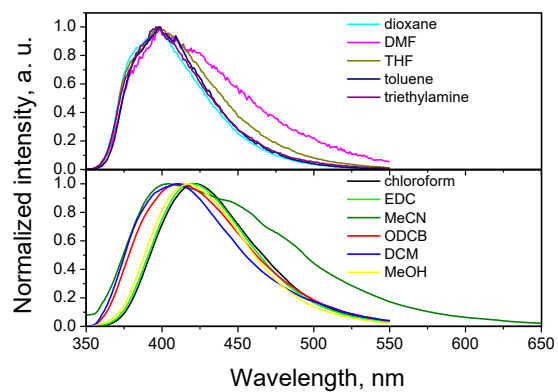


(b)

**Figure S3.** Absorption (a) and PL (b) spectra of **2** solutions

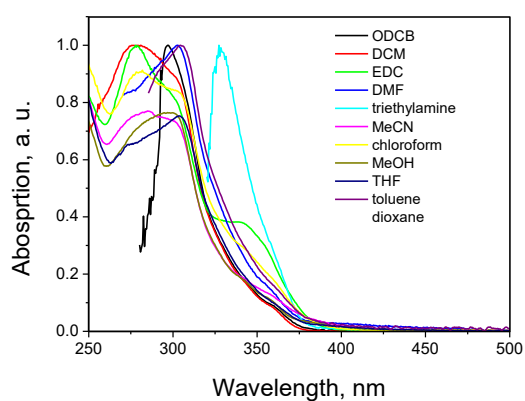


(a)

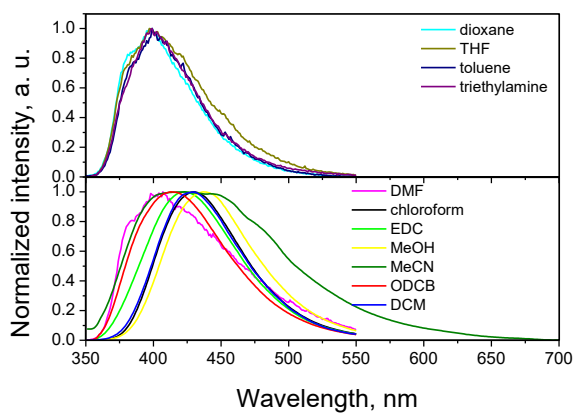


(b)

**Figure S4.** Absorption (a) and PL (b) spectra of **3** solutions

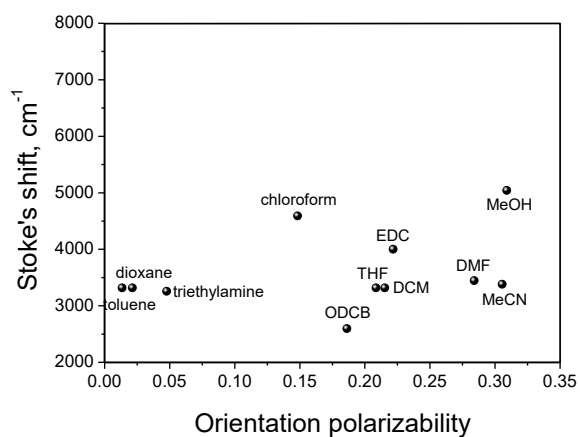


(a)

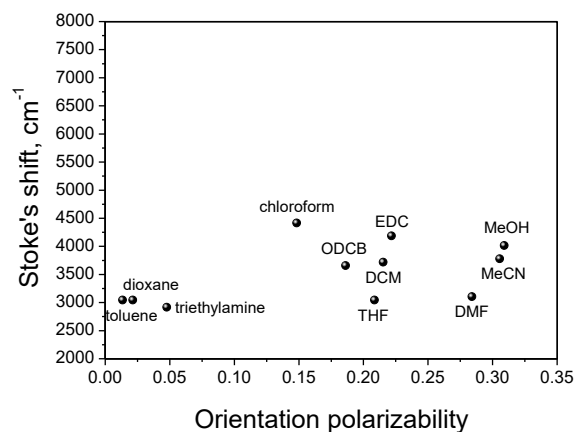


(b)

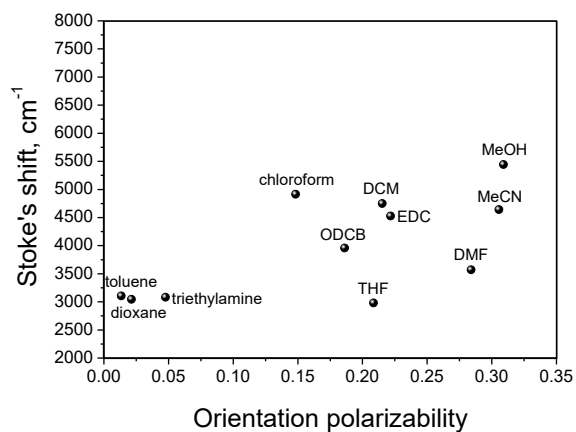
**Figure S5.** Absorption (a) and PL (b) spectra of **4** solutions



(a)

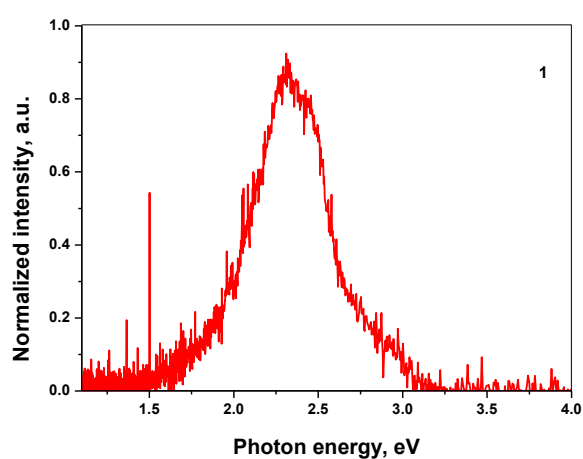


(b)

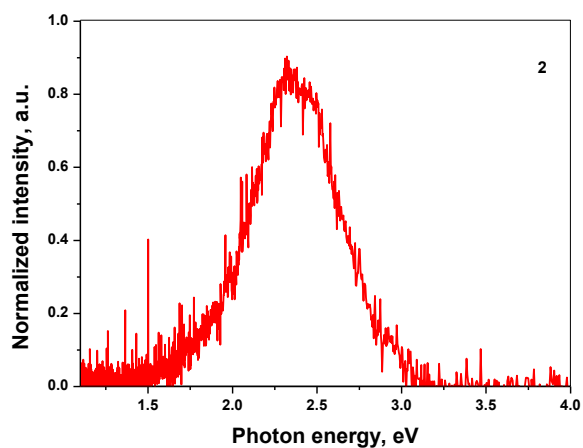


(c)

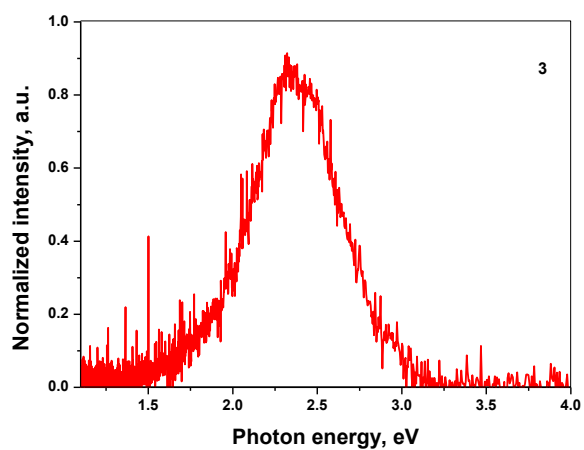
**Figure S6.** Lippert–Mataga plots for **1** (a), **3** (b) and **4** (c)



(b)

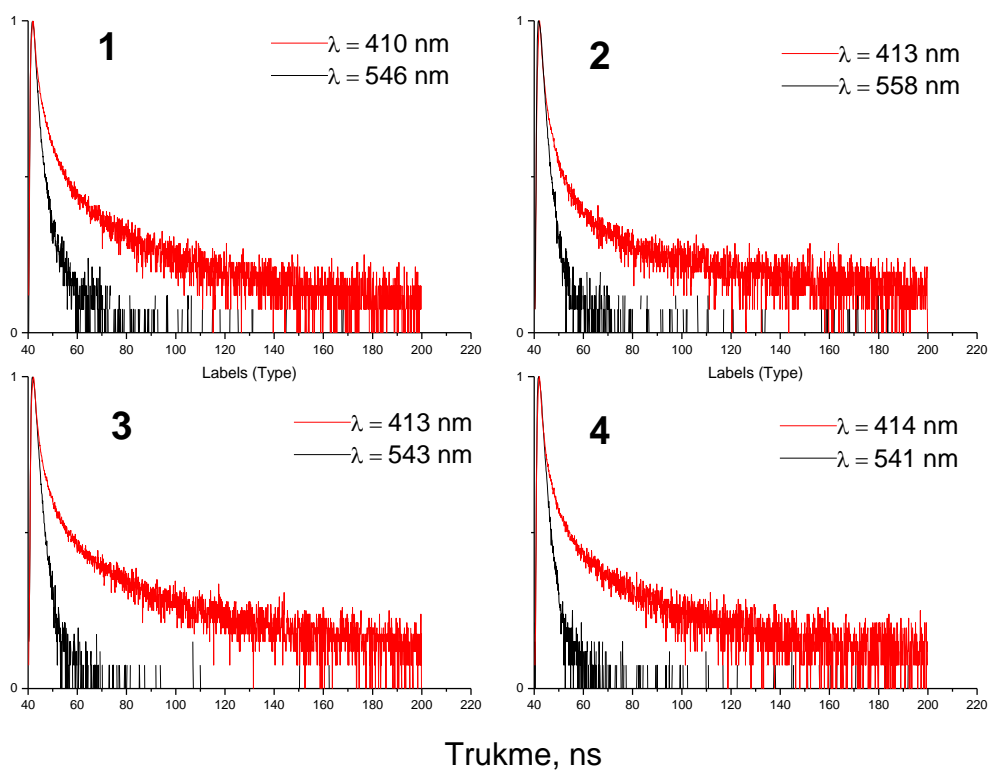


(b)

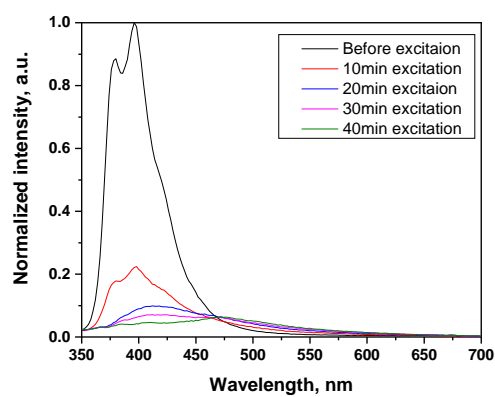


(c)

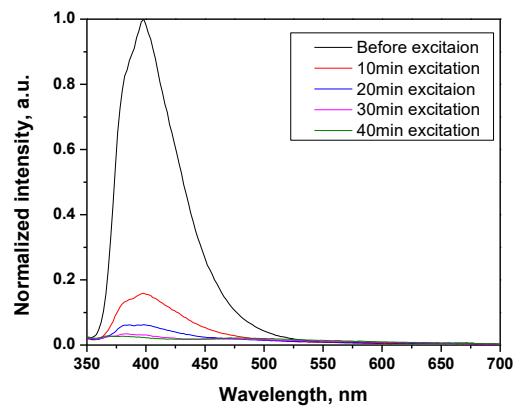
**Figure S7.** (a-c) Phosphorescence spectrum of dilute solutions of **1–3** in THF at 77K



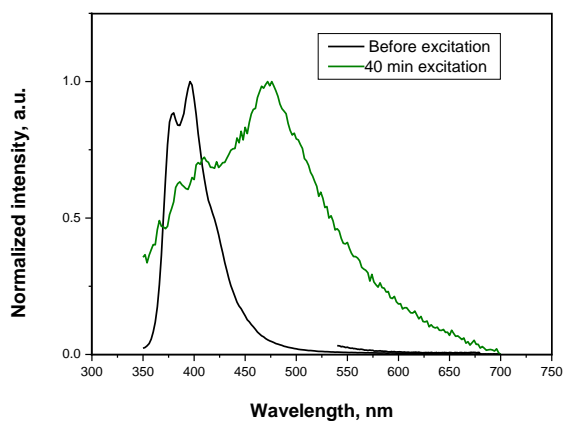
**Figure S8.** PL decay curves of solutions of **1–4** in THF



(a)

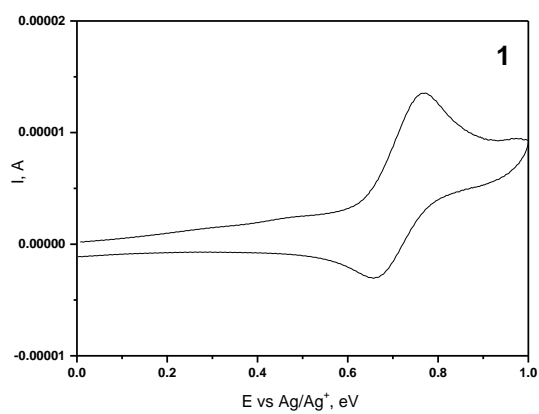


(b)

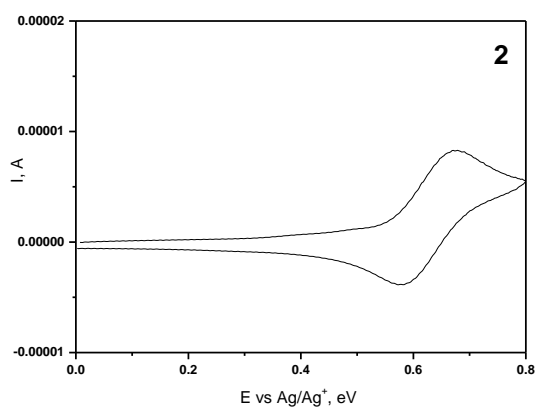


(c)

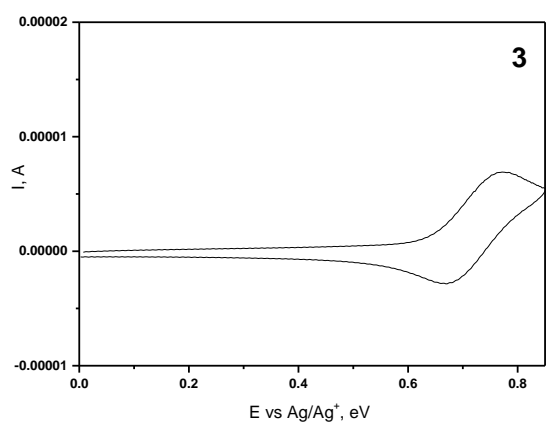
**Figure S9.** PL spectra of THF solutions of **1** (a) and **3** (b), normalized PL spectra of **3** (c) before and after 40 min photoexcitation by the UV light



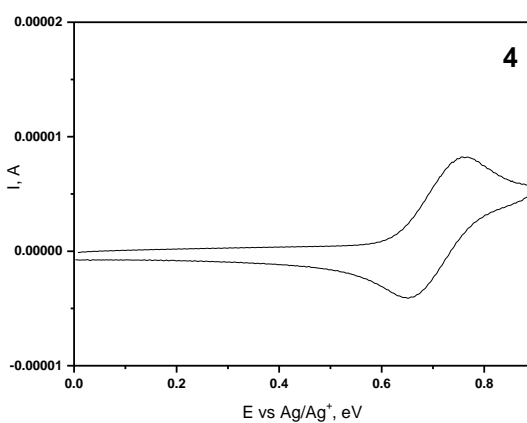
(a)



(b)

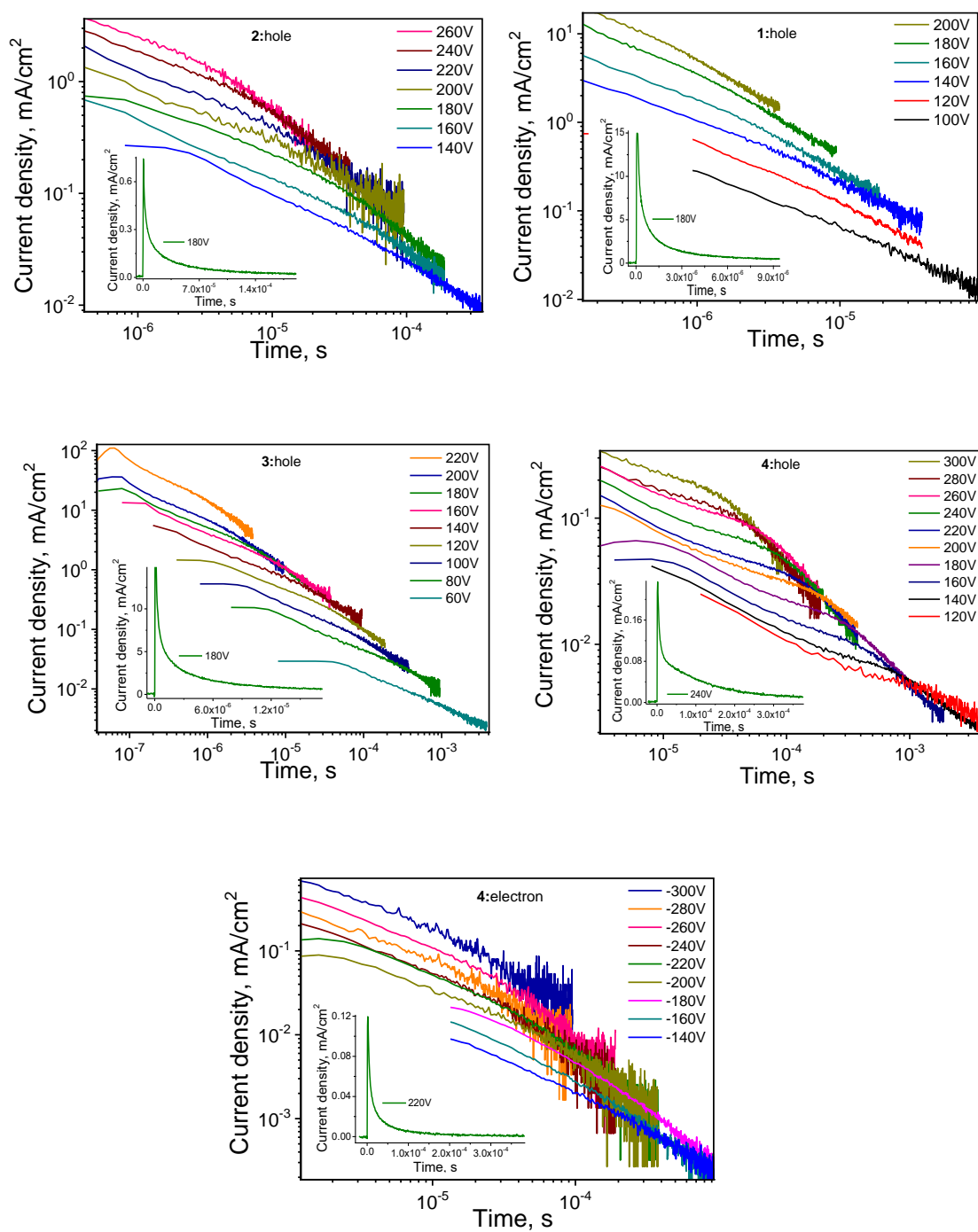


(c)

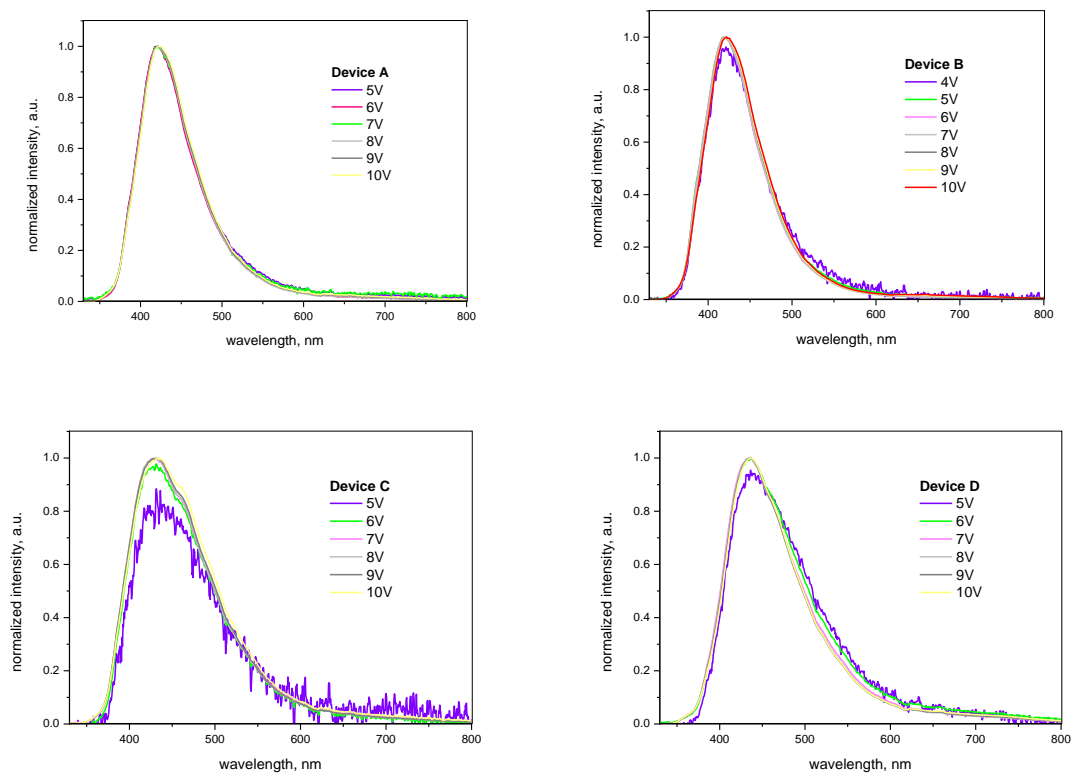


(d)

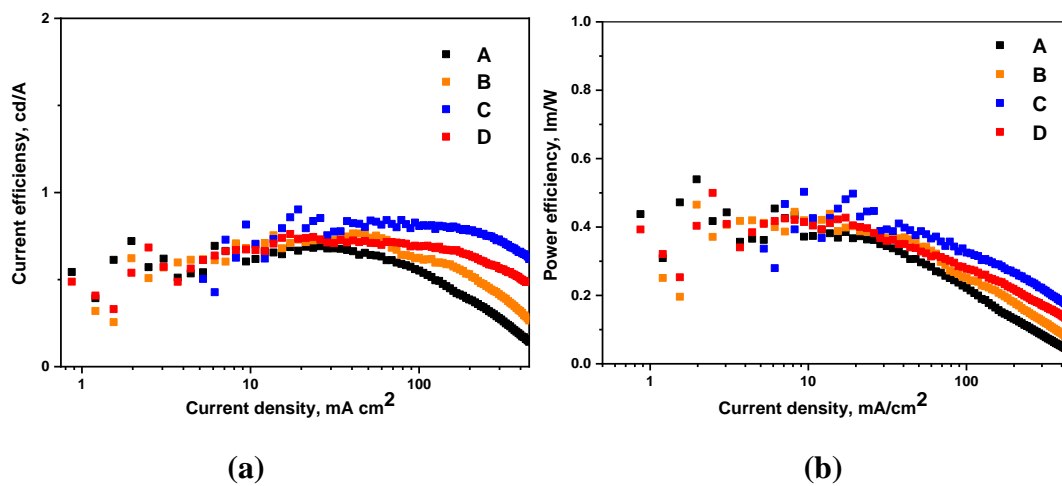
**Figure 10. (a-d) Cyclic voltammograms of compounds 1–4**



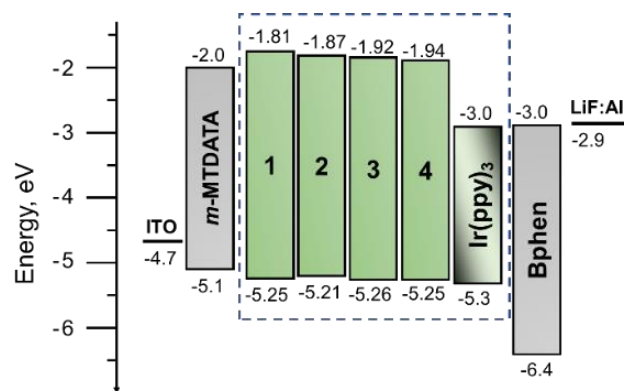
**Figure S11.** TOF signals for holes/electrons in vacuum-deposited layers of compounds 1–4 at different electric fields.



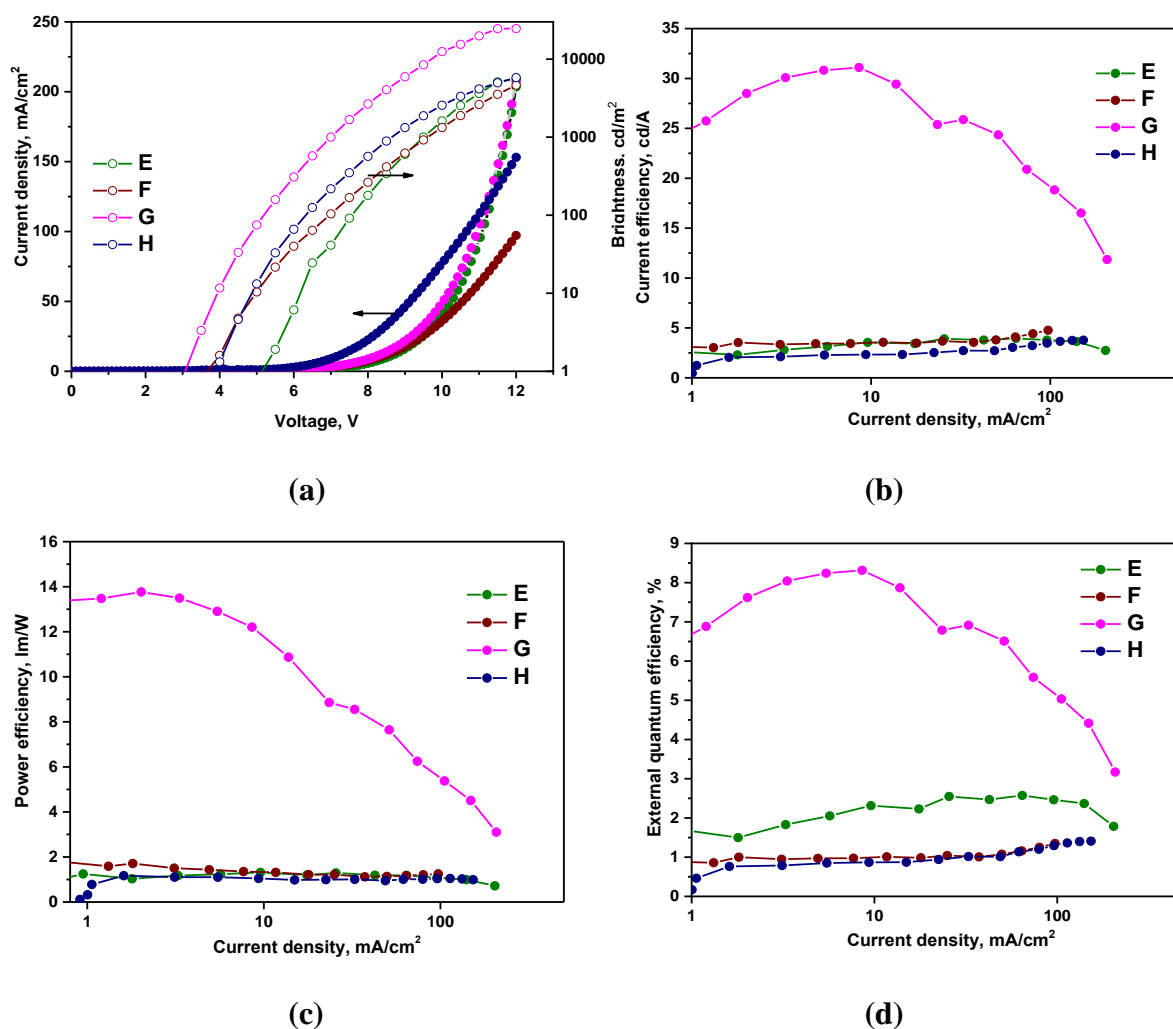
**Figure S12.** EL spectra of non-doped devices A–D at different voltages.



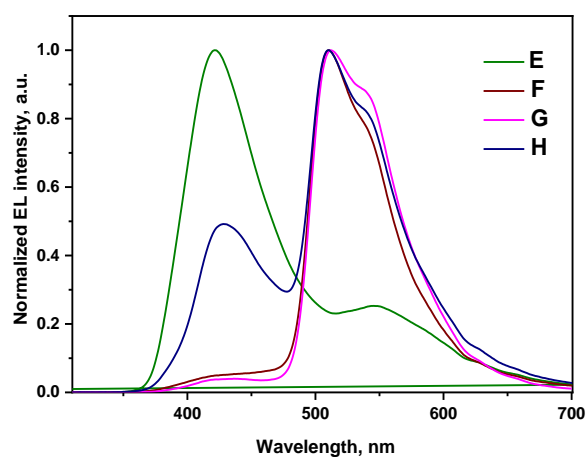
**Figure S13.** Electroluminescence characteristics of OLEDs A–D: **(a)** current efficiency versus current density, **(b)** power efficiency versus current density.



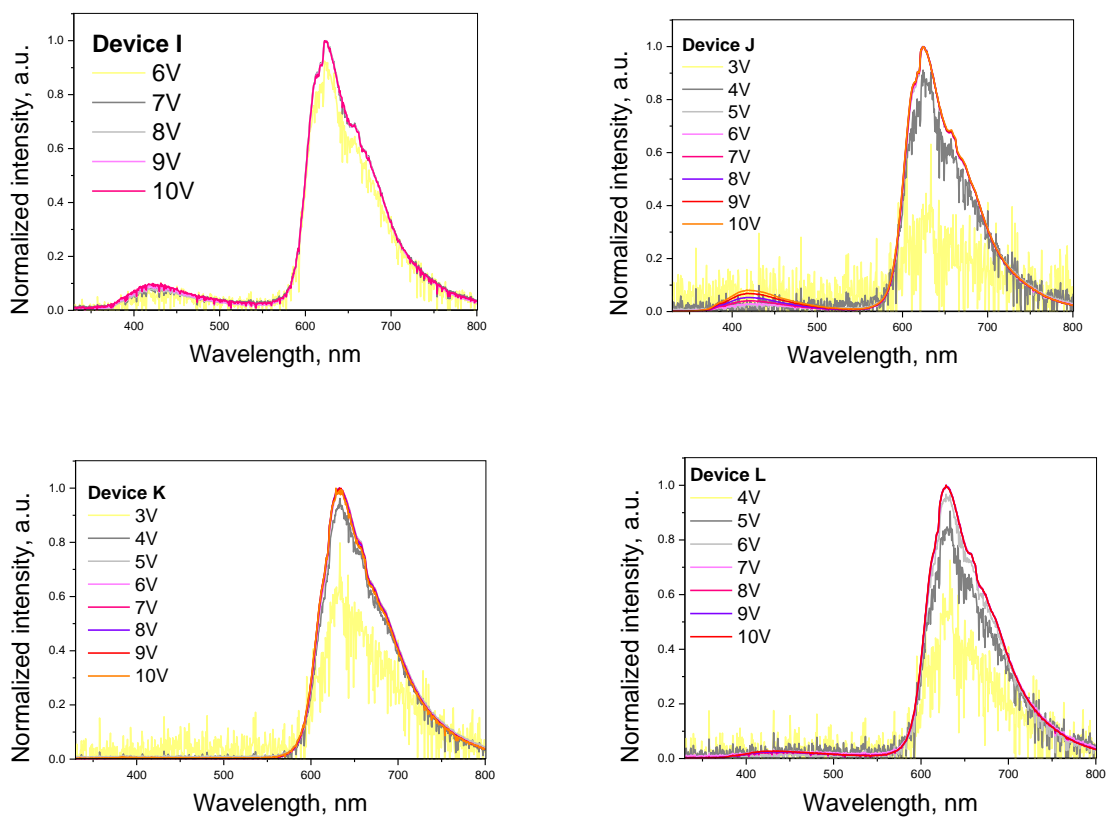
**Figure S14.** Diagram of energy levels of green PHOLEDs. 4,4',4''-tris[phenyl(m-tolyl)amino]triphenylamine (m-MTDATA) and bathophenanthroline (Bphen) were used as hole and electron transporting layers, respectively.



**Figure S15.** Electroluminescence characteristics of PHOLEDs **E–H**: (a) current density and luminance versus voltage, (b) current efficiency versus current density, (c) power efficiency versus current density, (d) external quantum efficiency versus current density



**Figure S16.** Electroluminescence spectra of PHOLEDs **E–H** recorded at applied voltage of 10 V.



**Figure S17.** EL spectra of devices **I–L** at different voltages.

## References

- [1] Miyamoto, E.; Yamaguchi, Y.; Yokoyama, M. Ionization Potential of Organic Pigment Film by Atmospheric Photoelectron Emission Analysis. *Electrophotography* **1989**, *28*, 364–370. <https://doi.org/10.11370/isjepj.28.364>.
- [2] Okamoto, S.; Tanaka, K.; Izumi, Y.; Adachi, H.; Yamaji, T.; Suzuki, T. Simple measurement of quantum efficiency in organic electroluminescent devices. *Jpn. J. Appl. Phy. Part 2 Lett.* **2001**, *40*, 783–784. <https://doi.org/10.1143/jjap.40.l783>.