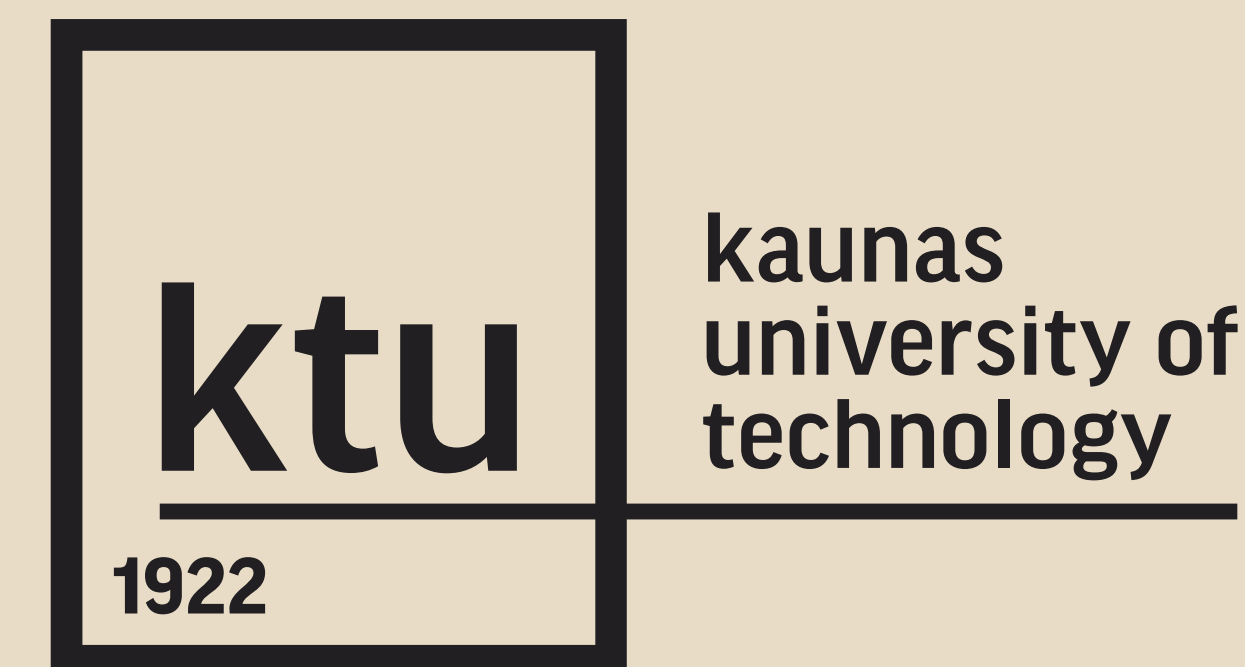


THERMALLY CROSS-LINKABLE HOLE-TRANSPORTING MATERIAL FOR EFFICIENT SOLUTION-PROCESSED ORGANIC LIGHT EMITTING DIODES



Deepak Kumar Dubey¹, Sujith Sudheendran Swayamprabha¹, Rohit Ashok Kumar Yadav¹, Daiva Tavgeniene², Raminta Beresneviciute², Greta Jacunskaitė², Nizy Sara Samuel², Dovydas Blazelevicius², Dmytro Volyniuk², Saulius Grigalevicius^{2*}, Jwo-Huei Jou¹

¹ Department of Materials Science and Engineering, National Tsing Hua University, Hsinchu, Taiwan

² Department of Polymer Chemistry and Technology, Kaunas University of Technology, Kaunas, Lithuania

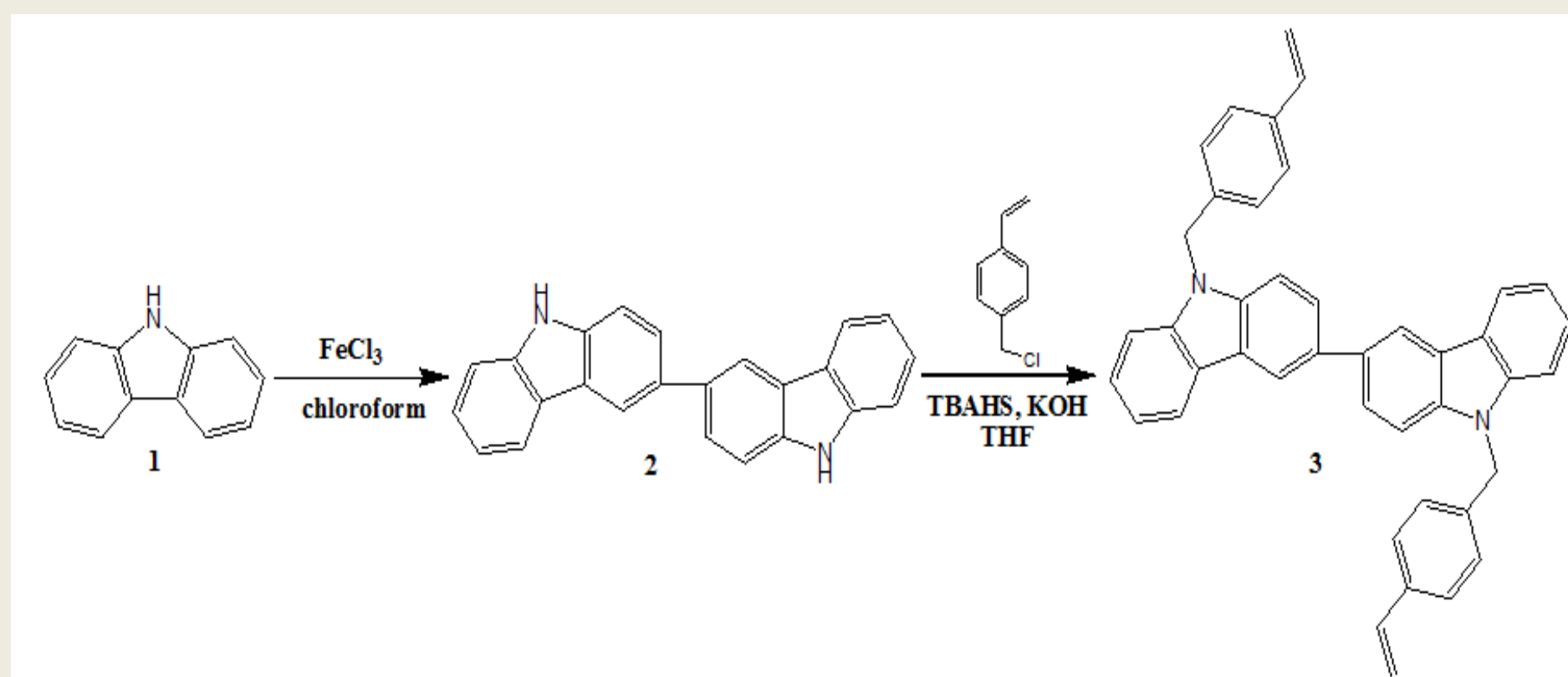
* saulius.grigalevicius@ktu.lt

Introduction

Organic light-emitting diodes (OLEDs) have drawn enormous attention in academia and industries because of their amazing applications in both next generation full-color flat-panel display and solid-state lighting, owing to their potential for fabrication over a large area, light weight, rapid response, low power consumption, and wide viewing angle. There are two general approaches to fabricate OLEDs, one is thermal evaporation process under high vacuum whereas another one is based on solution process feasible deposition. The latter one seems to be more superior and attracted by the academia and industry because of its descriptive features such flexible, roll-to-roll manufacturability on larger area and cost effective with high efficiency if one can prevent the blending or dissolution of formerly deposited layers throughout the consecutive deposition of multilayer stacks.

In this study, we have designed and synthesized a novel thermally cross-linkable HTL material, **VyPyMCz** for solution-process based multilayer monochromatic and white OLEDs. The compound **VyPyMCz** exhibits high triplet energy and cross-linking function due to the presence of a carbazolyl moiety and two vinylphenyl groups, respectively. OLEDs with the cross-linking layer show low operating voltages and improved power efficiencies as compared with the corresponding reference devices (without HTL). For example, the studied red, blue, and green devices showed an increment of 58%, 78% and 40% in power efficacy at 1,000 cd m⁻² and an increment of 75%, 30% and 20% in maximum luminance, respectively.

Synthesis



Scheme 1. Schematic illustration of the synthesis of carbazole based cross-linkable hole transporting material **VyPyMCz**.

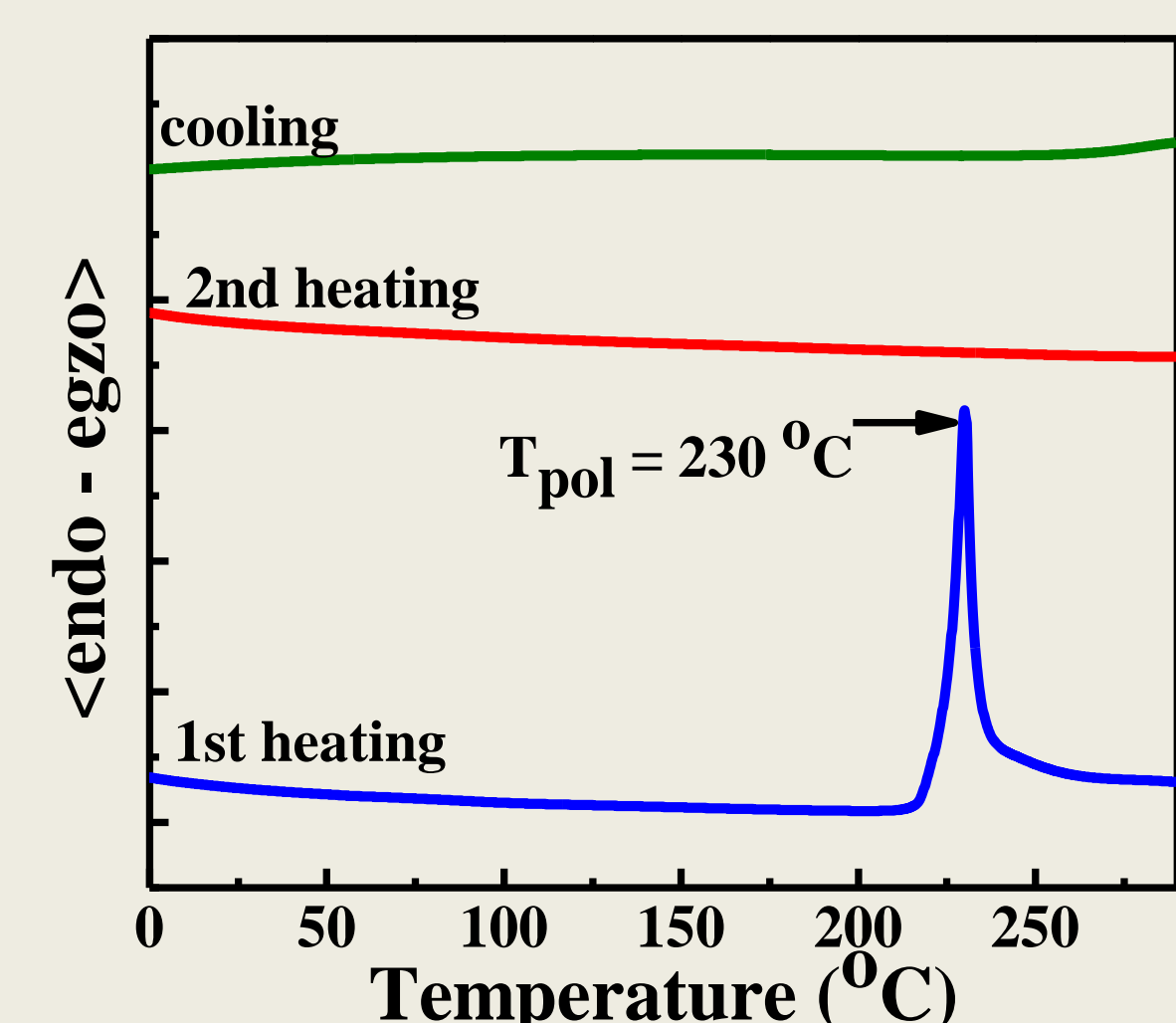


Figure 1. DSC curves of the material VyPyMCz, heating rate 10 °C/min.

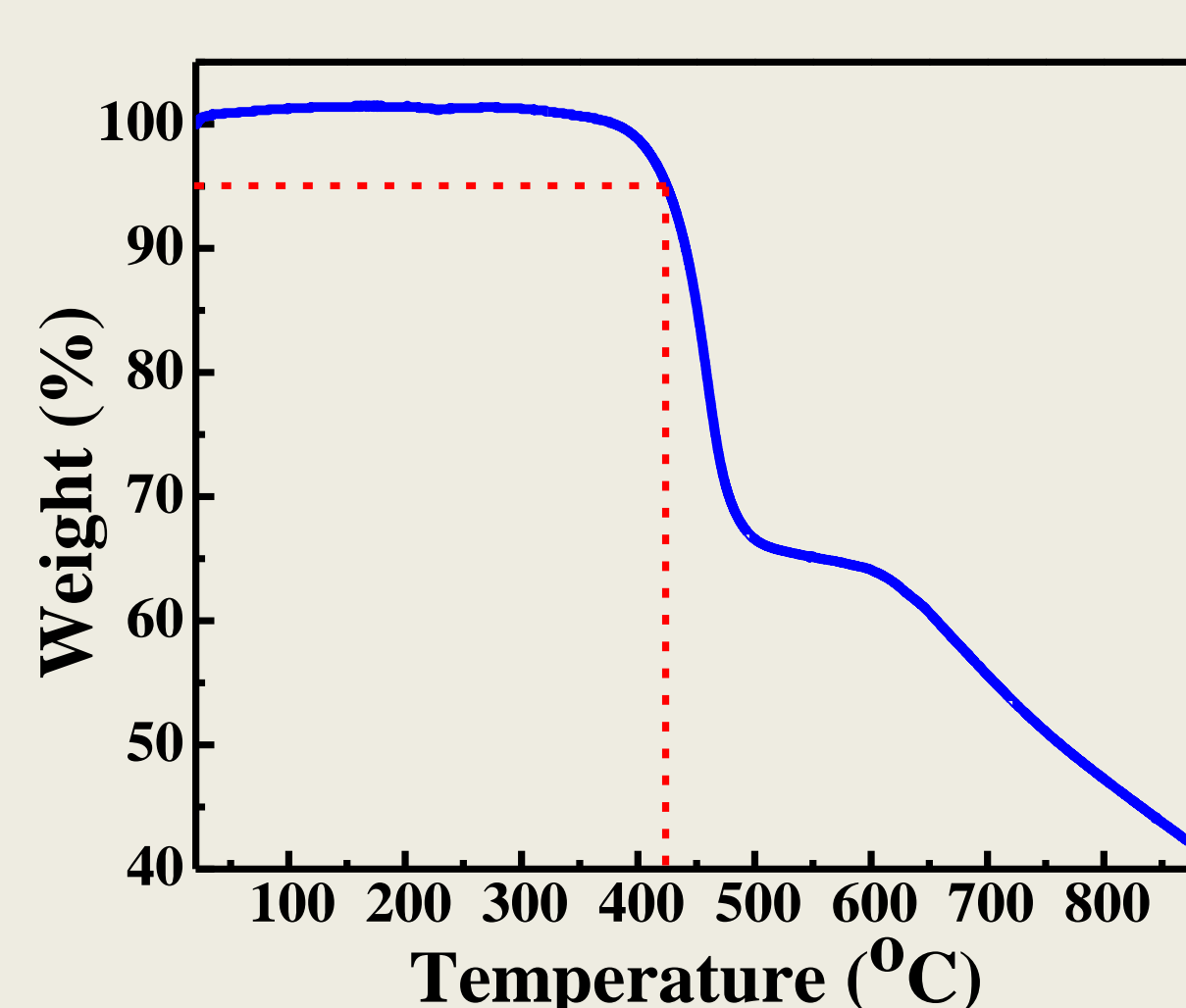


Figure 2. TGA curve of the material VyPyMCz, heating rate 10 °C/min.

Thermal, photophysical, and electrochemical characteristics

Compound	λ_{em}^a [nm]	T_g^b [°C]	T_d^c [°C]	μ_h^d [cm ² V ⁻¹ s ⁻¹]	HOMO ^e [eV]	LUMO ^e [eV]	HOMO ^f [eV]	HOMO ^f [eV]	Band Gap ^g [eV]	E _T ^f [eV]
VyPyMCz	402	60	423	3.6 x 10 ⁻⁴	-5.25	-1.87	-5.25	-1.25	3.38	2.87

^aEmission peak; ^bGlass transition temperature; ^cDecomposition temperature ^dHole mobility at the electric field of 3.3 x 10⁵ cm⁻¹; ^eHOMO and LUMO were calculated from the redox potentials measured by cyclic voltammetry (CV) method.; ^fCalculated by using B3LYP/6-31G (Density functional theory (DFT) and time-dependent DFT (TD-DFT)); ^gCalculated from its intersection point of the absorption and emission spectra.

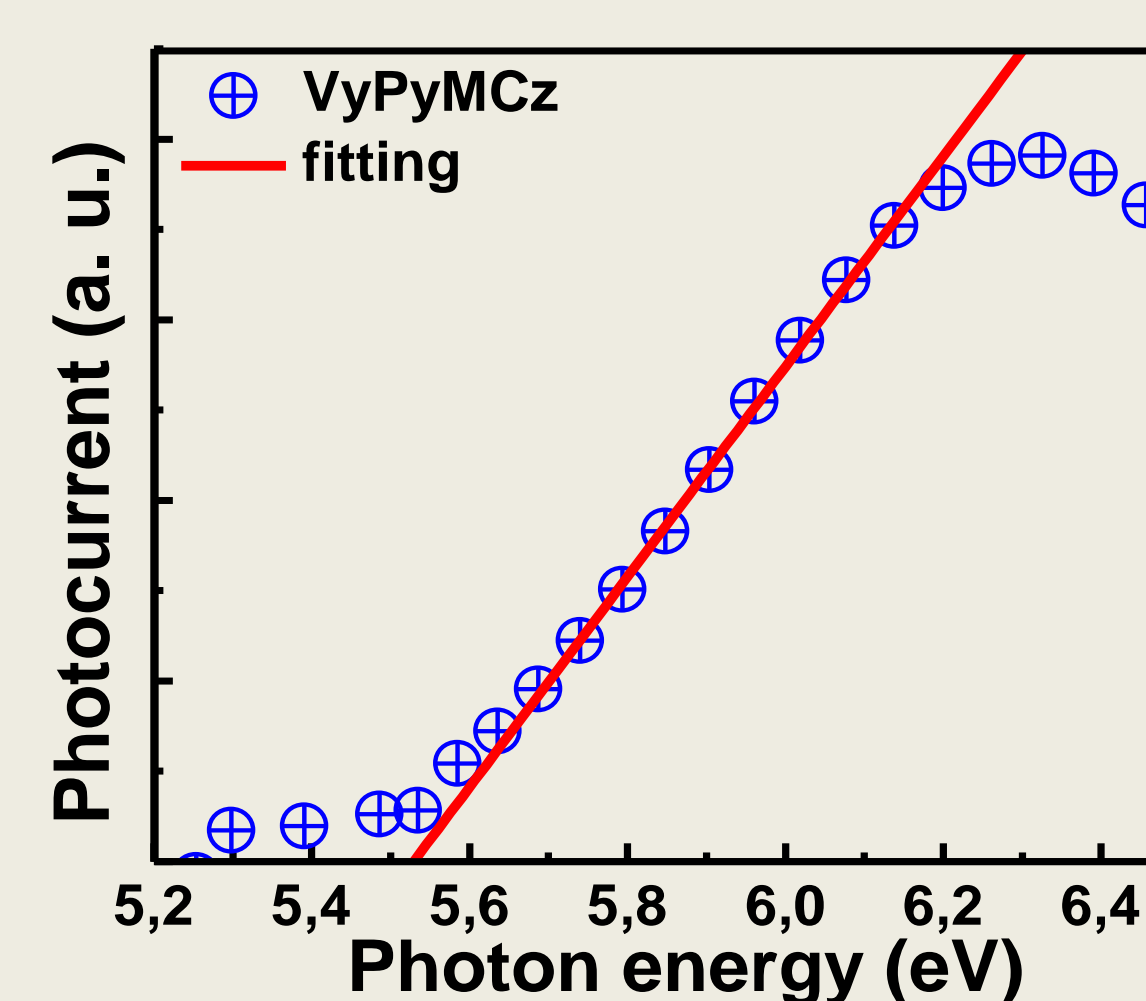


Figure 3. Electro-photoemission spectra of the VyPyMCz thin film.

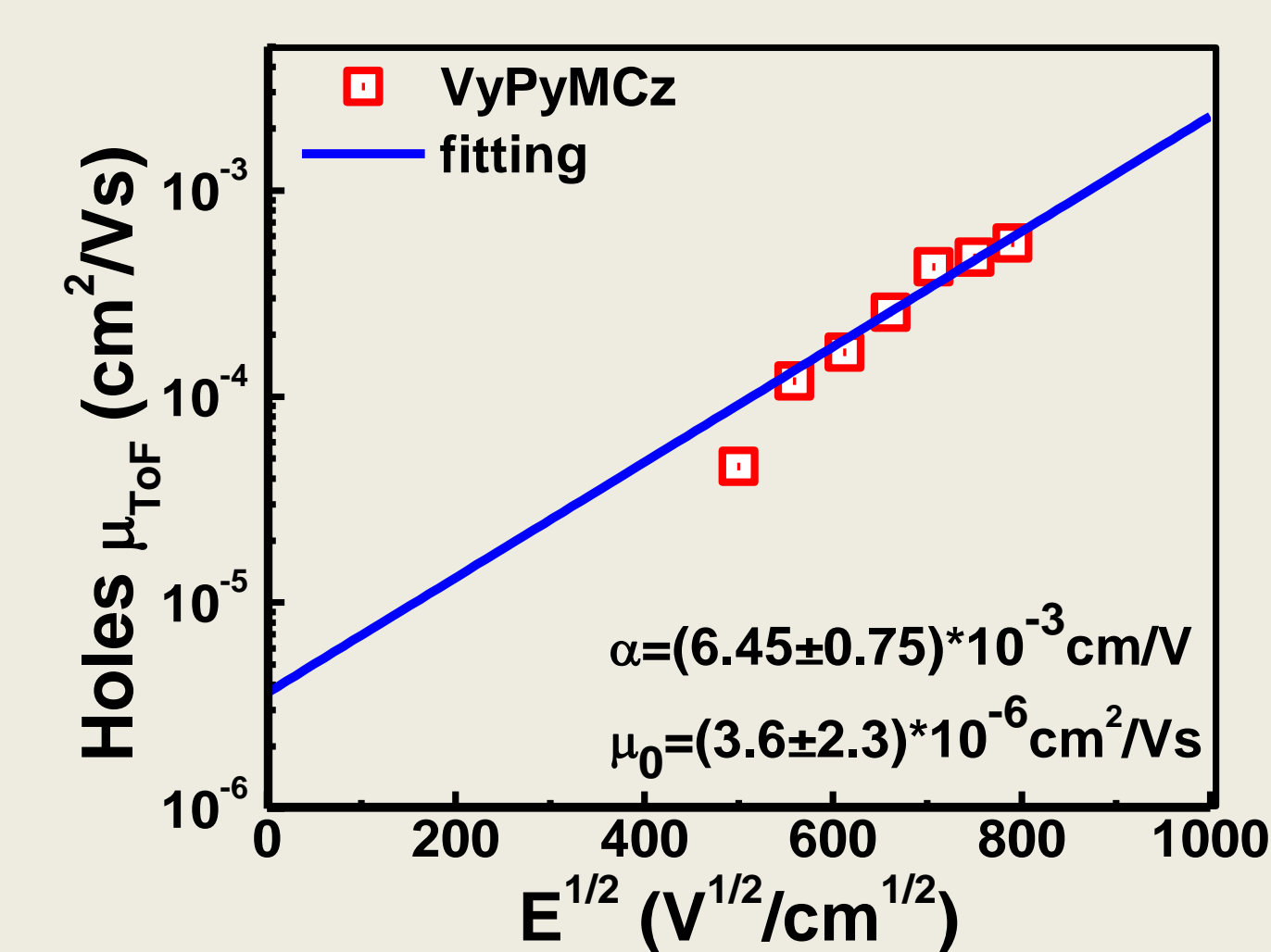


Figure 4. The hole mobility of the HTM VyPyMCz as a function of the square root of electric field.

OLED characterisation

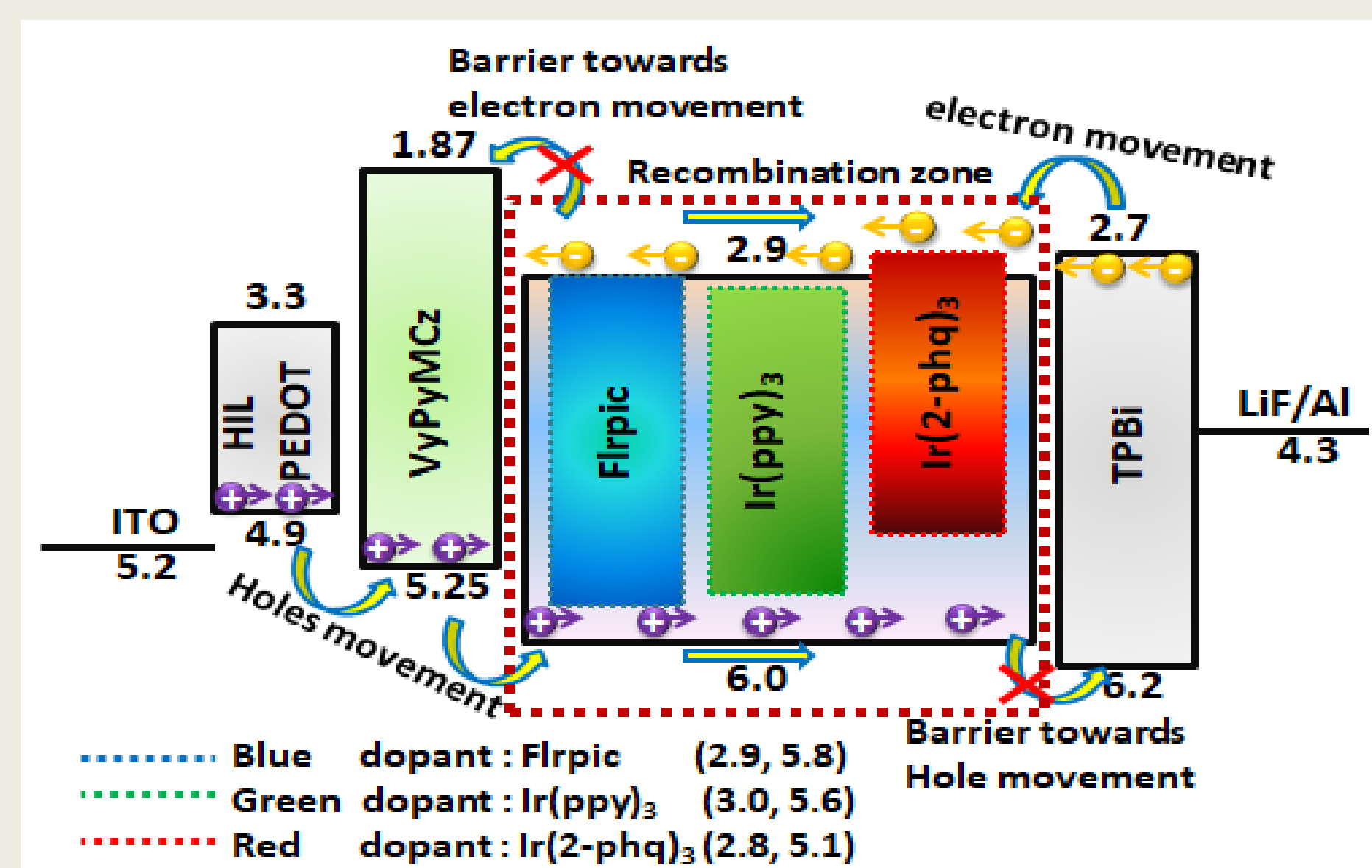


Figure 5. Schematic diagram of the energy levels of the fabricated solution-processed red, green, and blue OLED devices consisting of CBP and VyPyMCz as the host and cross-linkable hole transporting HTL, respectively.

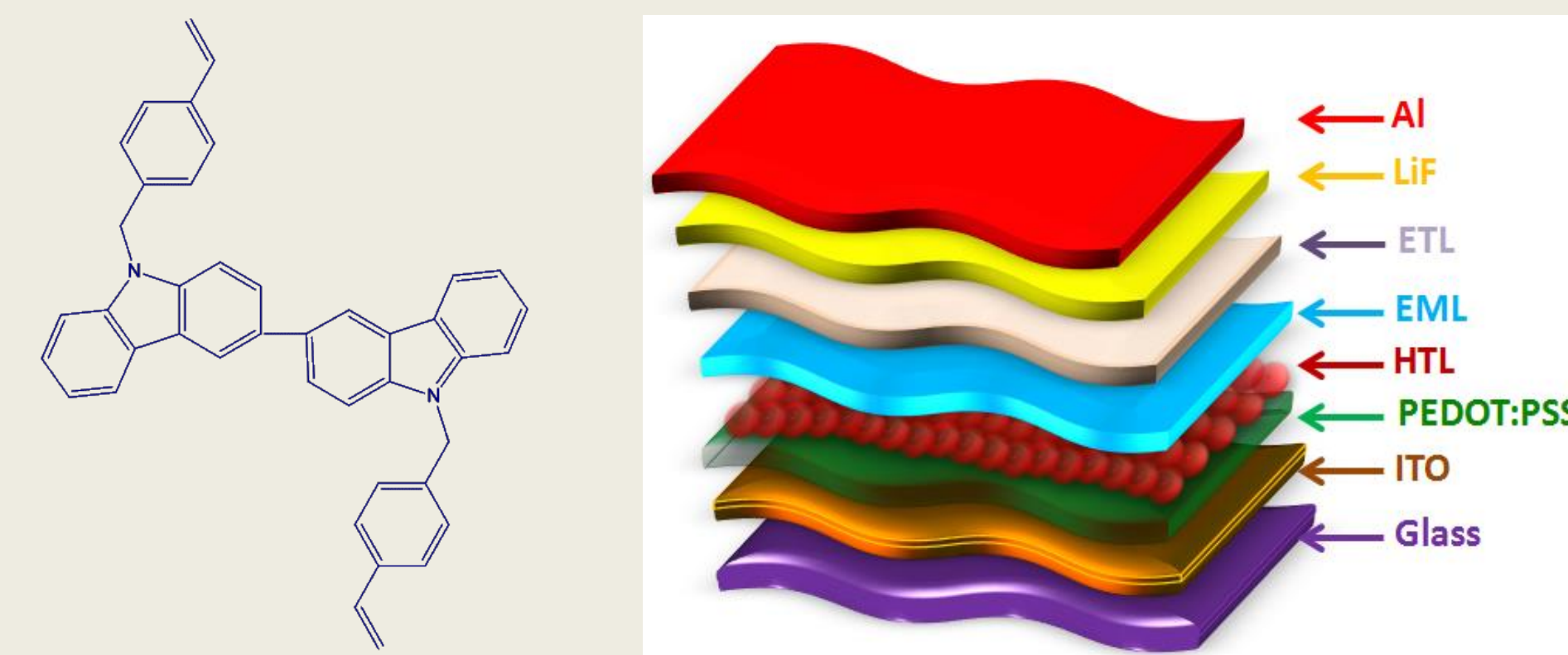


Figure 6. Chemical structure of novel synthesized cross-linkable hole transporting material (**VyPyMCz**), schematic illustration of OLEDs using **VyPyMCz** as a hole transporting layer.

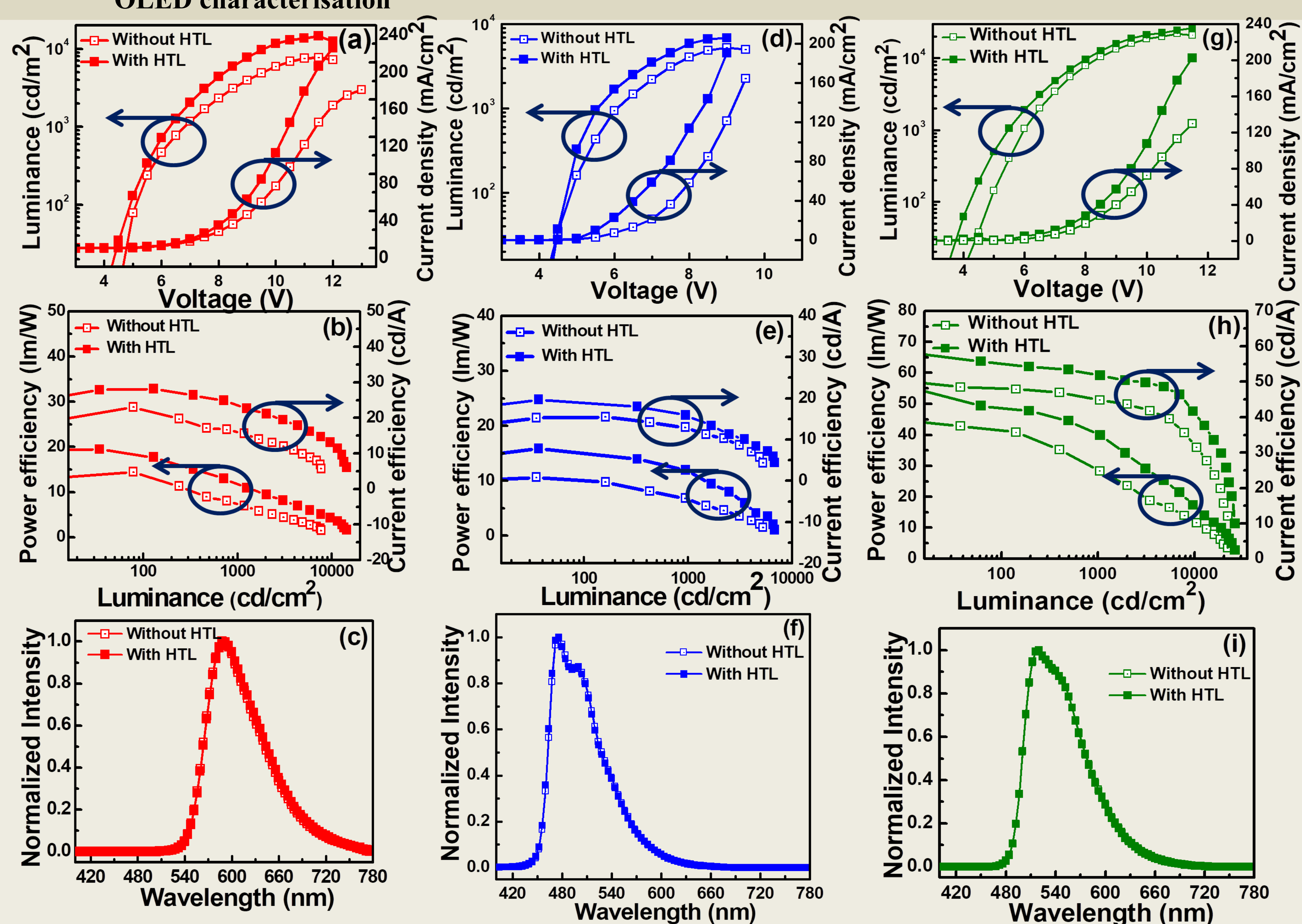


Figure 7. Effect of the presence and absence of the solution-processed cross-linkable hole transporting and electron confining layer, VyPyMCz, on the (1) luminance-voltage-current density, (2) power efficiency-luminance-current efficiency and (3) electroluminescent characteristics of the (a-c) red device, (c-f) blue device and (g-i) green device consisting of CBP as a host.

Conclusions

In this study, a novel class of solution process feasible and thermally cross-linkable hole-transporting material, **VyPyMCz**, has been designed, synthesized and successfully incorporated in the multilayered OLEDs. This compound features a larger bandgap and a higher triplet energy level. **VyPyMCz** can be thermally cross-linked to form a solvent-resistant layer upon isothermal heating at 250 °C for 30 min. The crosslinked **VyPyMCz** layer possesses not only hole-collecting ability (HOMO = -5.25 eV) but also electron-confining capability (LUMO = -1.87 eV), which significantly improve the number of exciton into the desired recombination zone, hence noticeable efficiency enhancement for red, green, and blue OLEDs, especially at higher voltage. The OLED device consisting of low band gap red emitter displays 58 and 45% increments in the PE and CE at 1,000 cd cm⁻², respectively, that is 78 and 47% for high band gap blue emitter. The approach presented herein could become a general method fabricate a highly efficient solution processed OLEDs by significantly injecting holes in the desired emissive layer and preventing the overflow of electrons into the non-radiative HIL.

Acknowledgement. OLED materials were developed in the frame of project funded by the Research Council of Lithuania (grant No. S-LLT-19-2) and by MOST (Taiwan).