

Supplementary Material

Bottom Contact Metal Oxide Interface Modification Improving the Efficiency of Organic Light Emitting Diodes

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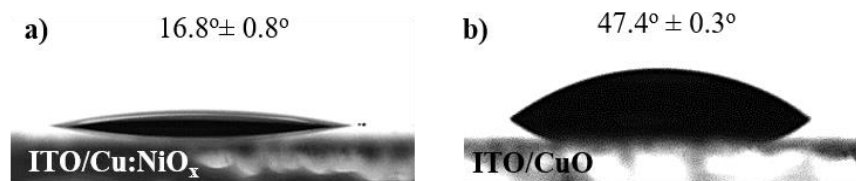


Figure S1. Static contact angle of deionized water droplet images on solution combustion synthesized (SCS) metal oxide substrates, a) ITO/Cu:NiO_x and b) ITO/CuO.

Table S1. Statistical analysis of electroluminescence performance of lab-scale 9 mm² organic light emitting (OLED) devices investigating the effect of spinel film thickness. Statistic sample size of 8 different devices was used to present average values, together with standard deviation, and maximum values in brackets.

Anode Electrode	Turn on voltage (at ~10 cd/m ²) [V]	Max. Luminance [cd/m ²]	Max. Current Efficiency [cd/A]	Max. Power Efficacy [lm/W]
ITO/PEDOT:PSS	2.6	9.0k ± 0.2k	4.9 ± 0.1 (5.0)	4.4 ± 0.1 (4.5)
ITO/(Cu-Li):NiCo ₂ O ₄ (~30 nm)	2.7	9.0k ± 0.3k	3.00 ± 0.4 (3.4)	2.5 ± 0.5 (2.8)
ITO/(Cu-Li):NiCo ₂ O ₄ (~20 nm)	2.7	8.9k ± 0.2k	3.2 ± 0.1 (3.4)	2.5 ± 0.3 (2.9)
ITO/(Cu-Li):NiCo ₂ O ₄ (~15 nm)	2.7	9.0k ± 0.5k	3.5 ± 0.1 (3.7)	2.9 ± 0.2 (3.1)
ITO/(Cu-Li):NiCo ₂ O ₄ (~10 nm)	2.8	8.0k ± 1.5k	3.4 ± 0.3 (3.7)	2.8 ± 0.3 (3.0)

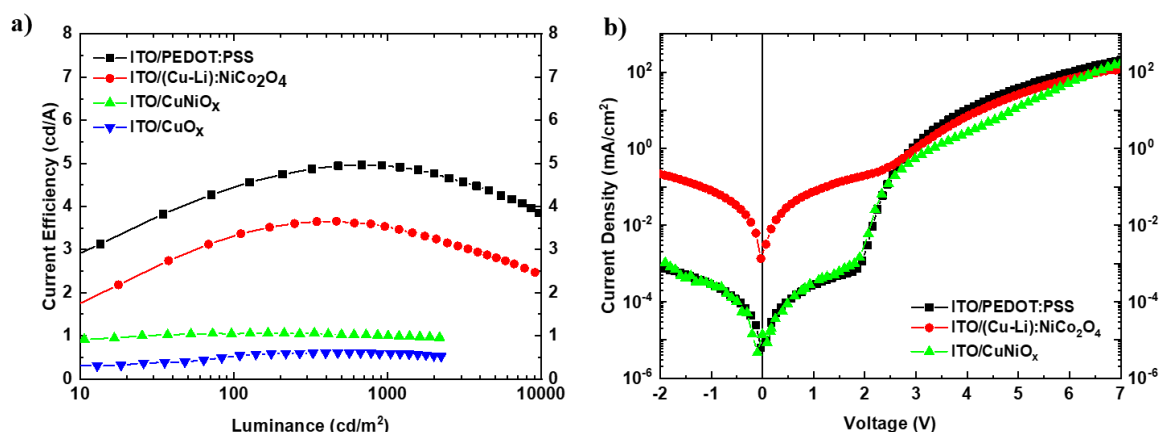


Figure S1. Performance of OLED devices based on ITO/SCS metal-oxide bottom electrodes, using different metal oxide interlayers, a) current efficiency over device luminance, b) semi-log JV - leakage current characteristics.

Replacing (Cu-Li):NiCo₂O₄ by Cu:NiO_x or CuO_x using a type B (indium tin oxide/metal oxide) device configuration, the performance analysis in Figure S2 points out to even lower efficiencies. Both Cu:NiO_x and CuO_x showed reduced performance in comparison to the optimized (Cu-Li):NiCo₂O₄, due to their much lower electrical conductivity Cu:NiO_x ($\sim 1.25 \times 10^{-3}$ S/cm) [1] and CuO_x ($\sim 1 \times 10^{-6}$ S/cm) [2], in comparison to (Cu-Li):NiCo₂O₄ (~ 4 S/cm) [3]. Studying the Figure S2b Cu:NiO_x presents similar leakage current characteristics to our reference device ITO/PEDOT:PSS due to its high LUMO level (~ 1.8 eV) of [4] that ensures electron blocking from super-yellow active layer, however the much lower forward bias current lead to reduced device efficiency. The effect of proper energy level alignment and electrical conductivity of MO, is strongly distinct in CuO_x with much deeper HOMO ~ 5.6 eV [5], strongly resisted the charge injection resulting in non-reproducible JVs , and significantly reduced device performance as shown in Figure S2a. Based on the fact that the low conductive PEDOT:PSS AI4083 formulation, used in this work has an electrical conductivity in the range of 10^{-3} – 10^{-4} S/cm, which is much lower than (Cu-Li):NiCo₂O₄ (~ 4 S/cm) we conclude that electrical conductivity plays a role among the examined MOs, however the proper energy level alignment with the active layer polymer is the most dominant parameter that affects the performance of OLED devices.

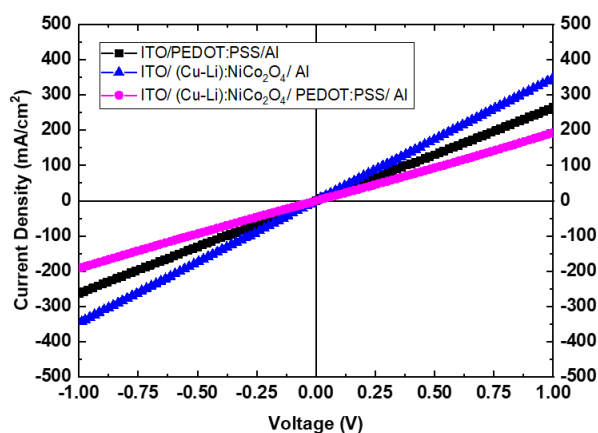


Figure S 2. Current density – voltage (JV) characteristics of PEDOT:PSS, (Cu-Li):NiCo₂O₄ and (Cu-Li):NiCo₂O₄/PEDOT:PSS, sandwiched between ITO anode and Al cathode electrodes.

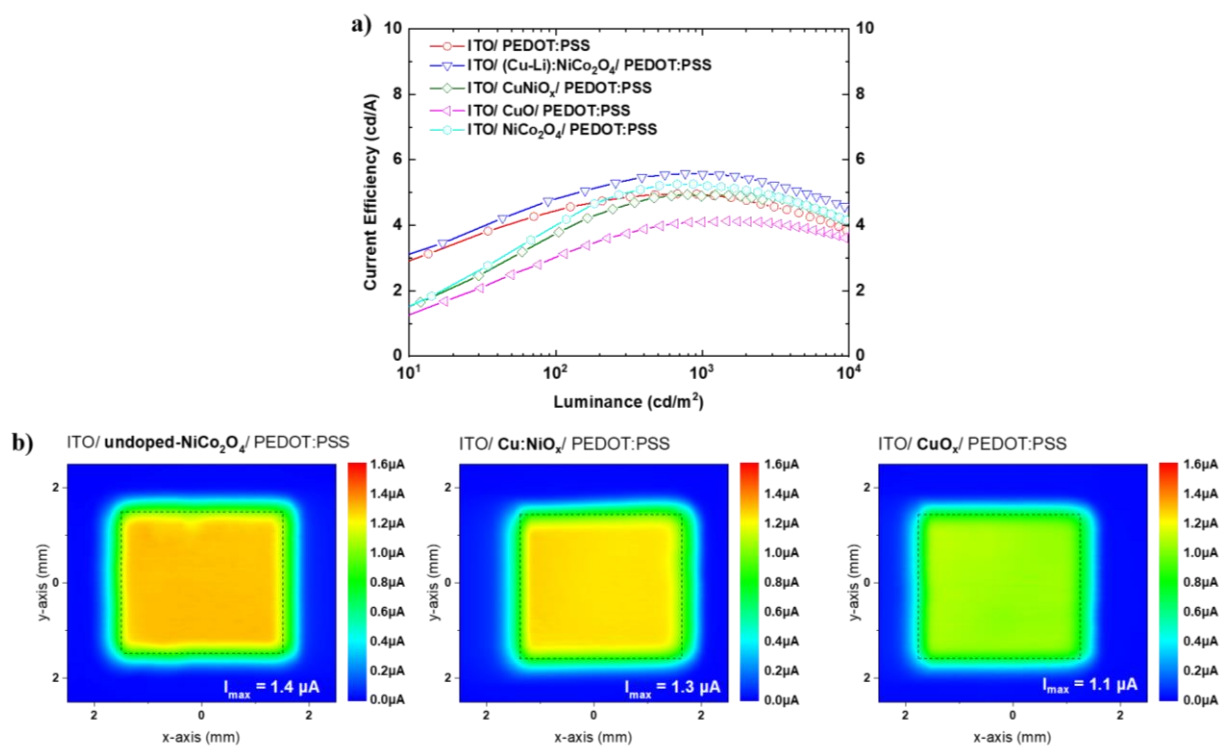


Figure S4. Performance of OLED devices based on ITO/SCS metal-oxide/ PEDOT:PSS bottom electrodes, using different metal oxide interlayers, a) current efficiency *versus* device luminance, b) photocurrent mapping images of complete 3 x 3 mm² SY OLED devices.

Device current efficiency presented in Figure S4a, follows the order, from highest to lowest, of ITO/undoped-NiCo₂O₄/PEDOT:PSS, followed by ITO/Cu:NiO_x/PEDOT:PSS and last ITO/CuO_x/PEDOT:PSS bottom electrode. The same trend was also observed in the generated photocurrent images Figure S4b, and the wetting properties of the examined SCS metal oxides (see Figure 1 and Figure S1).

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