

DOCTORAL THESIS

Solution-processable organic-inorganic hybrid transparent electrode for optoelectronic applications

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Abstract

The aim of this PhD thesis is to undertake a comprehensive research to study the optical, electrical, surface electronic and morphologic properties, formulation and surface modification of solution processable organic-inorganic hybrid transparent electrodes as well as their applications in optoelectronic devices. In this study, MoO₃ nanoparticles and graphene oxide (GO) nanosheets were incorporated into the poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate) (PEDOT:PSS) layer forming a hybrid anode interfacial layer (AIL) and subsequently a hybrid transparent electrode of AIL/silver nanowires (AgNWs), significantly improved charge injection in CdSe/ZnS-based quantum dot-light emitting diodes (QD-LEDs) and charge collection in bulk heterojunction (BHJ) organic solar cells (OSCs). The effect of oxidation behavior and charge transfer between PEDOT and MoO₃, as well as PEDOT and GO, on the enhancement in conductivity of hybrid PEDOT:PSS-MoO₃ and PEDOT:PSS-GO AILs was investigated systematically.

The presence of a PEDOT:PSS-MoO₃ AIL promotes a good interfacial contact between the hole transporting layer (HTL) and the solution-processed hybrid transparent electrode for efficient operation of QD-LEDs. This work reveals that the use of the hybrid PEDOT:PSS-MoO₃ AIL benefits the performance of QD-LEDs in two ways: (1) to assist in efficient hole injection, thereby improving luminous efficiency of QD-LEDs, and (2) to improve electron-hole current balance and suppression of interfacial defects at the QD/electrode interface. The surface wettability of the PEDOT:PSS-MoO₃ AIL was controlled

successfully for making a good contact between the HTL and the AgNWs, enabling efficient charge injection or charge collection, and thereby improvement in the device performance.

The effect of PEDOT:PSS-GO AIL on the performance of transparent QD-LEDs was also analyzed. The maximum brightness of the transparent QD-LEDs, made with a solution-processed hybrid top transparent electrode of PEDOT:PSS-GO/AgNWs, is 3633 cd/m² at 15 V, comparable to that of a structurally identical control QD-LED made with an evaporated Ag electrode, with a brightness of 4218 cd/m² operated under the same condition. The change in the hydrophobicity of the PEDOT:PSS-GO AIL, e.g., from the hydrophobic to hydrophilic characteristics, was observed. The interaction between PEDOT and GO nanosheets induces the transition between benzoid-quinoid structures, contributing to the enhanced charge carrier transport via the PEDOT:PSS-GO AIL. The energy level alignment at the HTL/electrode interface and the excellent electrical conductivity of PEDOT:PSS-GO/AgNWs transparent electrode result in an obvious improvement in the performance of QD-LEDs.

Transparent QD-LEDs also demonstrated remarkable efficiency via cathode interfacial engineering. Two cathode interfacial modifications include incorporating (1) a hybrid bathophenanthroline (Bphen):Cs₂CO₃-based electron transporting buffer layer (EBL) and (2) a conjugate polymer of poly[(9,9-bis(3'-((N,N-dimethyl)-N-ethylammonium)-propyl)-2,7-fluorene)-alt-2,7-(9,9-dioctylfluorene)] (PFN-Br)-based EBL. The approach of n-doping effect in the BPhen:Cs₂CO₃ EBL not only modifies the surface electronic properties of the ZnO electron transporting layer (ETL) but also improves the electron injection at the QD/cathode interface. The n-doping mechanism in the Bphen:Cs₂CO₃ EBL was investigated.

PFN-Br EBL has also been employed to tune the surface work function of ZnO ETL. It was observed that the ZnO/PFN-Br formed an interfacial dipole at the ETL/QD interface, which is suitable for efficient electron injection in the transparent QD-LEDs. In order to improve electron-hole current balance, a GO/MoO₃-based multilayer AIL was adopted facilitating efficient charge transfer through improved energy level alignment at the HTL/hybrid electrode interface. Photoelectron spectroscopy revealed tuned surface work function with reduced interfacial barrier for efficient hole injection in transparent QD-LEDs. In these devices, the cathode and anode interfacial modifications have been optimized and studied.

This study was also extended to investigate the effect of the organic-inorganic hybrid electrode on performance enhancement of all solution processable organic solar cells (OSCs). The reduction in series resistance and increase in shunt resistance of solution-processed OSCs originated from improved contact selectivity as well as enhanced charge collection efficiency. These properties are reflected in the significantly improved fill factor and short-circuit photocurrent density for the all solution-processed OSCs. Enhanced charge collection at the BHJ/electrode interfaces and improved process compatibility are mainly responsible for efficiency improvement in the cells. The outcomes of this work would allow further advances in device performance. This research also highlights the need to explore interfacial electronic properties and reduce energetic barrier at BHJ/electrode interfaces in fully solution-processed OSCs through photoelectron spectroscopy measurements. The results of this research demonstrate that the solution processable organic-inorganic hybrid transparent electrode developed in this work is beneficial for application in fully solution-processed optoelectronic devices.

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