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# A systematic approach to ZnO nanoparticle-assisted electron transport bilayer for high efficiency and stable perovskite solar cells

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ABSTRACT

Minimizing the interface loss of perovskite solar cells is critical to achieving high photovoltaic performance, and intensive research is underway on interfacial engineering in this regard. In this work, we introduce a ZnO nanoparticles (ZnO NPs) interlayer between phenyl-C61-butyric acid methyl ester (PCBM) and a metal electrode in order to reduce the interface loss due to charge recombination and device degradation, and also investigate the dependence of device performance on the thickness and morphology of the PCBM and PCBM/ZnO electron transport bilayer. After achieving optimized PCBM and ZnO thickness, the PCBM/ZnO bilayer-based devices reached an average power conversion efficiency of 15.63% (Max. 16.39%) with an open circuit voltage of 1.05 V, short circuit current density of 18.69 mA $cm^{-2}$, and fill factor of 79.95%. In addition, hysteresis behavior and atmospheric stability are significantly improved by the incorporation of a PCBM/ZnO bilayer. Therefore, the implementation of a PCBM/ZnO electron transport bilayer is a promising approach toward achieving a high-efficiency PSC with stable power output (low J-V hysteresis) and durability.



## 1. Introduction

In recent years, organic-inorganic hybrid perovskite materials with the chemical formula $CH_3NH_3PbX_3$ (X = I, Br, Cl) have attracted much attention because of their excellent photoelectric properties and high photovoltaic efficiency [1–6]. The organic-inorganic hybrid perovskite material has an appropriate band gap (1.5 eV), a high absorption coefficient, a long hole-electron diffusion length (100–1000 nm), and excellent charge-carrier mobility (~10 $cm^2 V^{-1}s^{-1}$); furthermore, the band gap and hole–electron diffusion length can be adjusted by mixing in various halides or by replacing the methylammonium organic species with other constituents such as formamidium and ethylammonium [7–13]. Interestingly, in a very short period of time, the power conversion efficiency (PCE) of perovskite solar cells (PSCs) has rapidly improved from 3.8% in 2009 to 23.7% in 2018 through intensive research on compositions, crystal growth control and morphologies, interfacial engineering, and device architecture [13–21].

PSCs with various device architectures have been explored, but recently, p-i-n type planar PSCs have begun to receive more attention [22–26]. This is because the mesoporous-structure and n-i-p type planar PSC exhibit good performance, but the formation of a mesoporous or compact $TiO_2$ layer requires high-temperature (>450 °C) processing; as a result, the potential for scale-up manufacturing and tandem and flexible substrate applications remains limited [13,22,27–30]. Further, the n–i–p type planar PSC with an n-$TiO_2$ as an electron transport layer exhibits a large hysteresis depending on the forward or reverse direction sweep [31,32]. In contrast, due to the passivation effect of fullerene derivative deposited on the perovskite, the p-i-n type planar PSC having the typical structure of a transparent conductive electrode/hole transport layer (HTL)/perovskite/electron transport layer (ETL)/metal electrode shows substantially reduced hysteresis behavior compared to the n-i-p type planar PSC [33–36].

Phenyl-C61-butyric acid methyl ester (PCBM) is commonly used as an ETL in PSCs of the p-i-n type planar structure, due to its high electron mobility and dissolution in non-polar solvents [22]. In addition, the lowest unoccupied molecular orbital (LUMO) energy level (3.9 eV) of PCBM can be well matched to the conductive band level of an organic-inorganic hybrid perovskite material, which allows for efficient electron transport between the organic-inorganic hybrid perovskite and PCBM interfaces [6,37,38]. However, it is difficult to form a uniform and defect-free PCBM film on

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