Green Synthesized NiO_x as the Hole Transport Layer for Inverted Perovskite Solar Cells

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1. Introduction: Since being discovered by Park in 2012 [1], perovskite solar cells (PSCs) have triggered rapid development and a power conversion efficiency (PCE) of 25.5%. Typically, inverted PSCs used PEDOT: PSS as the hole transport layer (HTL). However, this high cost and degradation of organic matter is a major concern for PSCs performance. Therefore, inorganic HTL materials such as NiO_x attract more attention due to superior hole mobility and outstanding stability. Additionally, NiO_x showed a favorable energy level with perovskite film and most importantly low-cost, and environment-friendly chemical synthesis capabilities. In this study, NiO_x particles from the green synthesis process have successfully tuned from micrometer-sized to nanoparticle size of ranged 10 nm, followed by preparing a smooth and compact NiO_x film and applied it as the HTL in inverted PSCs.

2. Materials and method: NiO_x powder was made by M. Akhtaruzzaman and colleagues [2]. NiO_x precursor solutions were prepared from NiO_x powders dissolved in nitric acid 3% and diluted with ethanol to 1 mL and spin-coated on FTO glass substrates. MAPbI₃ perovskite solution was prepared according to the procedure described by Y. H. Seo et al. [3]. Phenyl-C61-butyric acid methyl ester (PCBM) and bathocuproine (BCP) solution was spin-coated on the perovskite layer. Finally, 100-nm-thick Ag electrodes were deposited.

3. Results and Discussion:

 NiO_x particles powder obtained from the green synthesis process is shown in **Figure 1a**. The SEM image of NiO_x particles showed inhomogeneous and aggregated with particle size range 50 nm to several microns (**Figure 1b**). However, micrometer-sized NiO_x particles were difficult to attach to the FTO-substrate, subsequently produce incomplete surface coverage, which may lead to fewer electron blockages, and disrupts the device structure.

Therefore, NiO_x particles powder was further disperse using 3% nitric acid and diluted in ethanol solution. The NiO_x thin film exhibited dense, smooth, and full coverage with tiny NiO_x particles (**Figure 1c**). Ultimately, PSCs with NiO_x nanoparticles (NPs) HTL exhibited PCEs as high as 12.99%, which is superior

to those of PSCs with pristine NiO_x as the HTL (11.72%) (Figure 1d). The enhancement of J_{SC} and FF can be attributed to lowering the injection barrier at the interface between the dense NiO_x NPs thin film and perovskite, which facilitated efficient electron flow. The integrated values are coherent with the J_{SC} values extracted from the J–V curves (Figure 1e).



Figure 1. (a) NiO_x green synthesis process, (b) NiO_x particles from the green synthesis process before and (c) after reduced the particle size and deposited on FTO. Comparison of (d) *J-V* curves, (e) the IPCE spectrum of the best performing inverted PSCs using NiOx green synthesis with NiO_x reference.

We conclude that NiO_x green synthesized HTL showed smooth, dense, and exhibited the PCE of 12.99%.

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