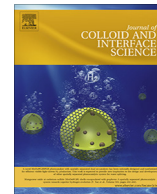




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Regular Article

Enhancing the performance of polymer solar cells using solution-processed copper doped nickel oxide nanoparticles as hole transport layer



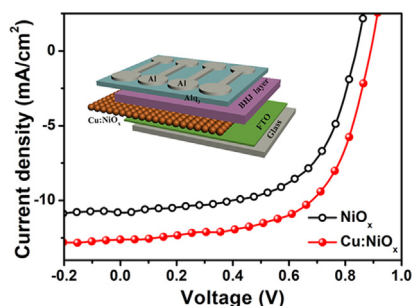
Shuai Huang^a, Yunhe Wang^a, Si Shen^a, Yuting Tang^a, Ancan Yu^a, Bonan Kang^{a,*}, S. Ravi P. Silva^b, Geyu Lu^a

^aState Key Laboratory on Integrated Optoelectronics, College of Electronic Science and Engineering, Jilin University, 2699 Qianjin Street, Changchun 130012, China

^bNanoelectronics Centre, Advanced Technology Institute, University of Surrey, Guildford, Surrey GU2 7XH, United Kingdom

GRAPHICAL ABSTRACT

Solution-processed Cu-doped NiO_x nanoparticles is an efficient hole transport layer in polymer solar cells. The Cu doping enhances the electrical conductivity and improves the interface contacts with the active layer, thereby improving the device efficiency.



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ABSTRACT

Polymer solar cells (PSCs) are considered promising energy power suppliers due to their light weight, printability, low-energy fabrication and roll-to-roll processability. Recently, the solution-processed NiO_x nanoparticles have been a desirable interfacial material for hole transport in the PSCs, instead of organic semiconductors. However, pure NiO_x films restrain the high performance of PSCs due to their poor electrical characteristics caused by the localized orbital distribution at the top of valence band. Therefore, metal ion doping has been explored as a method to endow NiO_x nanoparticles with the appropriate electrical characteristics. Herein, we applied solution-processed Cu-doped NiO_x (Cu:NiO_x) nanoparticles as an efficient hole transport layer (HTL) in PSCs. The Cu-doped NiO_x enhanced the electrical conductivity of the material and improved the interface contact with the active layer, which remarkably facilitated the hole extraction and effectively suppressed the carrier recombination at the interface. Thus, a higher power conversion efficiency of 7.05%, corresponding to an approximately 30% efficiency improvement compared with that of a pristine NiO_x interlayer (5.44%) in poly[N-9'-hepta-decanyl-2,7-carbazolealt-5,5-(4',7'-di-2-thienyl-2',1',3'-ben-zothiadiazole)]:[6,6]-phenyl-C₇₁-butyric acid methyl ester (PCDTBT:PC₇₁BM)-based PSCs, was achieved by the proposed device. The developed

* Corresponding author.

E-mail address: kangbn@jlu.edu.cn (B. Kang).

solution-processed Cu:NiO_x nanoparticles may be an excellent alternative for interfacial materials in PSCs or other optoelectronic devices requiring HTLs.

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1. Introduction

Polymer solar cells (PSCs) have attracted global attention from both the academia and industry on account of their interesting physical properties, which include light weight, printability, low-energy fabrication and roll-to-roll processability [1–5]. With the rapid development of polymer solar cells, they are gradually becoming a viable source of green renewable energy, and the power conversion efficiency (PCE) of device has been proven to exceed 11% for single-junction devices via interface engineering, material designing, and material designing [6]. Typical bulk-heterojunction (BHJ) PSCs are generally constructed with the active layer sandwiched between a low-work-function collecting electrode and an FTO substrate coated with hole transport layer (HTL) [7]. In order to obtain high-efficiency and stable organic photovoltaic cells, it is critical to tailor the active layer/electrode contacts, which can greatly influence the collection of free charge and the open circuit voltage (V_{oc}) of the device [8]. Various functional charge-transport layers must be incorporated between the photoactive layer and conductive electrode to suppress the charge carrier recombination and provide favorable charge transfer via ohmic contacts with the load connecting the external electrodes [9]. In general, for a material to be considered as an effective HTL in the photovoltaic cells, several electrical and optical requirements, such as large ionisation potential, low electrical resistance, good optical transparency, high electrical conductivity, and superior chemical stability, must be satisfied. In the typical architecture of PSCs, poly (3, 4-ethylenedioxythiophene): poly(styrenesulfonate) (PEDOT:PSS) is often used as an HTL due to its favourable solution processability, good conductivity and high work function (5.2 eV) [10]. The PEDOT:PSS interlayer can planarize the surface of indium tin oxide (ITO) electrode and enhance the charge extraction at the interface. However, the PSCs based on PEDOT:PSS HTL are generally limited by device performance problems and rapid degradation due to their long-standing acidic, hygroscopic characteristics and inability to effectively block electrons, both of which present a bottleneck in their commercialization [11]. Therefore, it is urgent to explore more effective and stable solution-processable anode buffer layers.

As a promising alternative to PEDOT:PSS, the solution-processed transition metal oxides with high work-functions and exhibiting facile synthesis, high optical transparency and good environmental stability have been widely investigated in the development of PSCs [12]. Until now, promising efficiency of PSCs have been achieved by incorporating the solution-processed molybdenum oxide (MoO₃), nickel oxide (NiO_x), vanadium oxide (V₂O₅), and tungsten oxide (WO₃) as the anode interfacial layer [13,14]. In contrast to *n*-type V₂O₅ and MoO₃, NiO_x is a promising alternative interfacial material for a *p*-type semiconducting hole harvesting metal layer adjacent to the ITO contacts in a number of photovoltaic systems due to its good optical transparency, high work function, excellent chemical stability, and superior hole extraction properties [15,16]. Thus, photovoltaic cells based on the NiO_x interlayers have been widely studied and developed to optimize their functions as the anode interlayers [17,18]. As the most successful example, Irwin et al. first introduce the pulsed laser deposited NiO film as the anode buffer layer in the P3HT:PCBM solar cells and achieve the enhanced photovoltaic performance in comparison with the cells with PEDOT:PSS interlayer

[19]. However, the vacuum-deposited fabrication process employed in the experiment has greatly increased the production cost of solar cells. After that, various approaches are applied to fabricate NiO_x thin films, such as electrochemical deposition [20], sol-gel process [21], sputtering deposition [22], atomic layer deposition [23], and Inkjet printing [24]. However, the photovoltaic performance of cells with the pristine NiO_x films are far from satisfied compared to the device with PEDOT:PSS interlayer and other organic HTLs. The reported PCE of the solar cells based on the PCDTBT:PC₇₁BM active layer is only 5.7% [25], which can be ascribed to their poor electrical characteristics caused by the localized orbital distribution at the top of valence band [26]. The low intrinsic conductivity of NiO_x will lead to the holes to accumulation near the BHJ layer interface, reducing the holes collection and enhancing the carrier recombination.

Therefore, it is imperative to explore some novel strategies, such as improvement of the quality of deposited NiO_x or doping the NiO_x, to increase conductivity and charge collection, thereby improving the device efficiency. Metal ion doping is considered as an effective approach to enhance the performance of photovoltaic cells. The film quality, conductivity, and surface roughness can be improved or adjusted by the substitution of Cu onto the NiO site [27,28]. In this work, we show that the device performance can be improved by incorporating the solution-processed Cu-doped NiO_x nanoparticles as the anode interfacial layer. Our proposed solution-processed Cu:NiO_x films show increased the hole mobility, electrical conductivity, and charge extraction, as well as suppressed charge carrier recombination. The proposed Cu-doped NiO_x nanoparticles produced remarkable improvements in power conversion efficiency (PCE) (7.05%) in PCDTBT:PC₇₁BM-based PSCs, which outperformed the devices with pristine NiO_x films (PCE 5.44%). This work demonstrates that appropriate Cu doping of NiO_x films has proved to be an efficient and simple approach for enhancing device performance and could be a promising material for other photoelectric devices where functional charge-transport layers are required.

2. Experimental

2.1. Materials and solutions

PCDTBT and PC₇₁BM were purchased from Rieke Metals. To obtain photoactive layer materials, PCDTBT (7 mg) and PC₇₁BM (28 mg) were dissolved at a weight ratio of 1:4 were dissolved in 1, 2-dichlorobenzene and stirred in a N₂ atmosphere. A facile non-toxic solution-processable process was conducted to prepare the undoped and Cu-doped NiO_x precursor solutions. Briefly, Ni (CH₃COO)₂·4H₂O and different quantities of Cu(CH₃COO)₂·H₂O were completely dissolved in a mixture of absolute ethanol and monoethanolamine to yield a deep green and homogeneous solution.

2.2. Solar cell fabrication

The pre-patterned FTO coated glass was successively rinsed with acetone, deionized water, and isopropanol for 15 min under sonication, dried at 100 °C for 10 min in an oven and then treated under O₂ plasma for 60 s. The pure and Cu-doped precursor solutions were spun coated onto the FTO at 1000 rpm for 30 s in ambi-

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