Niobium doped zinc oxide nanorods as an electron transport layer for high-performance inverted polymer solar cells

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Abstract

The wet chemical synthesis, characterization and applications of Nb doped ZnO nanorods in bulk heterojunction inverted polymer solar cells are presented in this paper. Zn_{1-x}Nb_xO with x ranging from 0.01 to 0.07 were successfully synthesized using a novel facile solution-processed wet chemical method. The structural and optical properties of the undoped and Nb doped ZnO nanorods were investigated by transmission electron microscopy, X-ray diffraction, UV–visible near infrared, and photoluminescence (PL) spectroscopies. A clear red shift was observed in the absorption spectra of Nb doped samples compared to the undoped ZnO nanorods. The defect emission peak in the PL spectrum of the Nb doped nanorods completely disappeared at the highest dopant concentration of x = 0.07. These undoped and Nb doped ZnO nanorods, in amalgamation with PTB7-Th:PC_{71}BM, were used in the inverted polymer solar cells as an electron transport layer which gives better power conversion efficiencies compared to the pure nanorods.

1. Introduction

Solar energy is an important candidate for the future energy market due to its unlimited supply, and its inexpensive, renewable, and clean nature [1]. For harvesting solar energy, organic photovoltaics seem to be a good option because of their excellent features including low cost, wide choice of materials, and low processing temperature [2]. Still, the reported power conversion efficiency of organic photovoltaics is very low because of the energy loss during the charge transfer process at the interface, and the poor charge transport properties of the conjugated...
polymers [3]. The grouping of poly (3-hexylthiophene) (P3HT) with fullerene derivatives has upgraded the power conversion efficiency of organic solar cells. This further unlocks the possibility of large-scale device manufacturing of thin-film organic solar cells [4]. In thin-film organic solar cells, the device construction and the selective active layer usually play a vital role in increasing the device efficiency. In order to meet these requirements, the inverted device architecture was presented to improve the stability of the interface. Moreover, it can decrease the active layer degradation and maximize the device performance [5–9]. Generally, glass substrates coated with transparent conductive metallic oxide are used for the fabrication of inverted polymer solar cells. Specifically, the word inverted is used to mention the flow of the charges in the reverse direction. For example under ambient conditions, it shows excellent environmental stability of the device due to the elimination of a low work function cathode in addition to a corrosive hole transport buffer layer known as PEDOT:PSS. To obtain stable solar cell devices, the buffer layer plays a pivotal role in the preparation of organic polymer solar cells [10]. Zinc oxide (ZnO) is used as an electron transport layer in organic polymer solar cells due to its excellent transport capabilities [11–13]. Moreover, the conduction band of ZnO matches the lowest unoccupied molecular orbital (LUMO) level of most acceptors. Scientists have done a remarkable job at improving ZnO properties and performance as an electron transport layer [10,14,15]. An alternative tool towards improving the properties of ZnO as an electron transport layer has been achieved via insertion of a foreign atom in the ZnO lattice. Kim et al. [16] presented an efficient and stable inverted organic solar cell using lithium doping of ZnO to achieve a maximum efficiency of 5.49%. The production of organic solar cells using indium doped ZnO as electron transport layer [17] and PCDTB:PC71BM as the active layer revealed better power conversion efficiencies. Moreover, Aluminum, Boron, Gallium were some of the important dopant elements that have also positively contributed towards improving device performance [18,19]. Olson et al. [20] demonstrated the great effect of band gap tuning via Mg doping of ZnO in hybrid solar cells on obtaining the highest efficiencies in the bulk heterojunction configuration. Lu et al. [21] investigated Au/LiF modified ZnO interlayer to maximize the charge efficiency and obtained a 40% increase in power conversion efficiency. Moreover, Jiang et al. [22] used novel Nb-doped (0 0 1) - dominated anatase TiO2 nanosheets as photoelectrodes in dye-sensitized solar cells thus achieving 10% power conversion efficiency, which is higher than the undoped counterpart. There is also a recent report on the simultaneous doping of Sr, Nb in TiO2 particles, which improve the efficiency of the short circuit current density of the dye-sensitized solar cells [23]. To the best of our knowledge, there are few scientific reports on niobium doped ZnO. For instance, Satheesan et al. [24] determined room temperature ferromagnetism in Mn and Nb co-doped ZnO nanoparticles prepared by the sol-gel method. For gas sensing applications, Kim et al. [25] designed Nb doped ZnO nanowalls by radio-frequency (RF) magnetron sputtering at 250 °C. Lin et al. [26] prepared Nb doped ZnO thin films on glass substrates using pulsed laser deposition (PLD) and investigated the effects of substrate temperature by analyzing the structural, optical, and electrical properties. Doping of Nb into ZnO is favorable because of the difference of the three valence electrons between Nb5+ and Zn2+. Thus each Nb atom can donate three valence electrons to the overall electrical conductivity. Here we report a wet chemical solution method for the synthesis of undoped and Nb doped ZnO nanorods, modified from the Pacholski method [27]. Methanol was employed as a solvent. The dopant concentration x ranging from 0.01 to 0.07, where x = 0.01 (1%), 0.03 (3%), 0.05 (5%) and 0.07 (7%) doped samples. A 0.1 M solution of zinc acetate and Niobium powder at the desired concentration were dissolved in 200 ml of methanol, then heated up to 60 °C. 1.5 ml of deionized water was then added to the reaction mixture. The 0.1 M solution of sodium hydroxide was then dissolved in 100 ml of methanol by ultrasonication in a separate flask. Subsequently it was added to the former solution (reaction mixture) drop by drop under argon atmosphere with a constant magnetic stirring at 60 °C. The development of the reaction was observed by the changes in the color of the reaction mixture i.e. upon the addition of NaOH, the solution becomes milky, after 30 min it transforms into a transparent solution, and in the end it becomes milky once again. After 2 h and 15 min, undoped and Nb doped ZnO nanoparticles were obtained. The reaction was stopped and the solution rested overnight until the precipitates settles down to the bottom. White precipitates were obtained for the pure ZnO nanoparticles and grayish black precipitates for Nb doped ZnO nanoparticles. The precipitates were further washed several times with DI water and ethanol. The following chemical reactions [28] match well with our experimental findings.

$$Zn^{2+} \text{(solution)} + 2OH^- \rightarrow Zn(OH)_2^{2-} \text{(Solid)}$$ (1)

$$Zn(OH)_2^{2-} \text{(Solid)} + 2OH^- \rightarrow ZnO_2^{2-} \text{(Solution)} + 2H_2O$$ (2)

$$ZnO_2^{2-} \text{(Solution)} + H_2O \rightarrow ZnO \text{(Solid)} + 2OH^-$$ (3)

For the synthesis of undoped and Nb doped ZnO nanorods, the nanoparticle solution was concentrated using a rotary evaporator under reduced pressure at 60 °C until the solution is colorless. Afterwards, this solution was placed in an oil bath at 60 °C with constant magnetic stirring. The reaction was stopped after 55 h and the nanorods were allowed to settle down to the bottom. The white and grayish black solid precipitates consisting of undoped and Nb doped ZnO nanorods were washed several times with ethanol and deionized water. These thoroughly washed undoped and Nb doped ZnO nanorods were stored for further characterization.

2.2. Fabrication of bulk heterojunction inverted polymer solar cell

PTB7-Th and PC71BM were purchased from 1-Material, Solarmer Material, and Solenne BV, respectively. Our detailed fabrication process for the reference bulk heterojunction inverted polymer solar cell devices based on ZnO as an electron transport layer has been previously reported [29,30]. Initially, cleaning of the ITO-coated glass substrate with a sheet resistance of 10 Ω cm−1 (purchased from Shenzhen Nan Bo Group, China) was achieved by ultrasonication in detergent, deionized water, acetone, and isopropanol for 20 min each, then dried in an oven at 60 °C overnight. Afterwards, Nb doped ZnO nanorods were dissolved in (1:1 v/v) methanol and chloroform, at 15 mg/ml. A thin layer of nanorods layer is that it exhibits better contact with the organic active layer due to its hydrophobic nature. Finally incorporating the optimized electron transport layer with a structure of ITO/Zn1−x NbO/PC71BM:PTB7-Th/MoO3/Ag exhibits preferable power conversion efficiencies exceeding 7% (up to 7.39%).
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