

# Enhanced photocurrent and stability of organic solar cells using solution-based NiO interfacial layer

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## Abstract

Metal oxide semiconductors are promising interfacial materials for organic photovoltaics (OPVs) because of their electrical properties and solution processability. In this article, we report the fabrication of poly(3-hexylthiophene):[6,6]-phenyl-C<sub>61</sub> butyric acid methyl ester (P3HT:PCBM) OPV devices incorporating solution-based NiO interfacial layers that show promising enhancements of the device photocurrent and stability. We discuss the impact of parasitic shunt and series resistances on device performance as well as the ambient degradation of these devices, studied with intensity modulated photocurrent spectroscopy (IMPS). The results showed that charge extraction was predominantly affected by degradation via decrease in carrier mobility and increased trapping/recombination, revealing the physical mechanism behind the degradation observed.

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**Keywords:** Solution process; Nickel oxide; Parasitic resistances; Intensity modulated photocurrent spectroscopy (IMPS)

## 1. Introduction

Organic photovoltaics (OPVs) have attracted significant research interests due to their potential for large-scale, flexible and lightweight applications via high throughput and inexpensive solution-based manufacture (Dennler et al., 2007; Pagliaro et al., 2008; Kalowekamo and Baker, 2009; Pivrikas et al., 2011; Weickert et al., 2011). A critical component in OPVs is the charge selective interfacial layer, which has been shown to have a significant influence on the device performance and stability (Shrotriya et al., 2006; Yamanari et al., 2010). The conventional OPV structure currently employs the standard poly(3,4-ethylenedioxythio-

phene):poly(styrenesulfonate) (PEDOT:PSS) hole selective interfacial layer. However, there is sufficient evidence to suggest that PEDOT:PSS plays a significant role in causing device degradation (de Jong et al., 2000; Kemerink et al., 2004; Sharma et al., 2011; Voroshazi et al., 2011), resulting in considerable efforts to develop suitable replacements for PEDOT:PSS.

Amongst the various alternatives developed, metal oxide semiconductors are promising due to their ideal optical and electrical properties, as well as their ambient stability (Shrotriya et al., 2006; Jung et al., 2011; Kinoshita et al., 2008; Steim et al., 2010; Tao et al., 2009). Of the several metal oxides available, NiO is known to enhance device performance and stability (Betancur et al., 2011; Irwin et al., 2008; Park et al., 2010; Steirer et al., 2010, 2011). However, the bulk of these studies using NiO involved vacuum deposition processes, which are neither scalable nor cost effective. Hence, there have been attempts to solution process NiO to keep

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its fabrication in line with scalable printing techniques. Notably, Steirer et al. demonstrated solution processing for NiO films with a proprietary Ni-ink precursor (Steirer et al. 2010, 2011). More, however, can be done to exploit conveniently accessible Ni sources and characterize such interfacial layers using relevant techniques. In particular, intensity modulated photocurrent spectroscopy (IMPS) is a powerful, non-destructive characterization technique that is commonly used for the analysis of various optoelectronic devices. Recently, IMPS has been applied to OPV devices (Byers et al., 2011; Chen et al., 2009; DiCarmino and Semnikhin, 2008) to obtain information on carrier transport and recombination/relaxation processes.

In this article, we discuss the performance of poly(3-hexylthiophene):[6,6]-phenyl-C<sub>61</sub> butyric acid methyl ester (P3HT:PCBM) solar cells incorporating a solution processed NiO interfacial layer. The synthesis of the NiO interfacial layers is facile, inexpensive and uses nickel acetate, which is commercially available as the precursor. This procedure is also not energy intensive when compared to conventional vapour deposition methods for the fabrication of such thin films (sputtering, evaporation, etc.). We discuss the effects of parasitic shunt ( $R_{sh}$ ) and series ( $R_s$ ) resistances on device performance. The NiO interfacial layer was observed to enhance device photocurrent and stability compared to reference PEDOT:PSS devices. The effect of ambient degradation on charge transport and recombination processes in the device was studied by IMPS. It is concluded that the loss in photocurrent observed during ambient degradation was due primarily to a reduction in carrier mobility as well as increased recombination and trapping.

## 2. Experimental section

NiO films were prepared by spin coating a precursor solution of nickel acetate tetrahydrate ( $\text{Ni}(\text{CH}_3\text{COO})_2 \cdot 4\text{H}_2\text{O}$ ) (Alfa Aesar) on pre-cleaned ITO (tin-doped indium oxide) glass substrates (20  $\Omega/\text{sq}$ , Xinyan Technology Ltd.). The substrates were cleaned with detergent followed by successive sonication baths of 15 min each in de-ionised water, acetone and iso-propanol (IPA), then finally dried at 60 °C in an oven. The substrates were treated with UV-ozone for 20 min before use. The precursor solution consisted of  $\text{Ni}(\text{CH}_3\text{COO})_2 \cdot 4\text{H}_2\text{O}$  dissolved in methanol with an equimolar amount of diethanolamine. The concentration of the precursor solution was varied between 0.1 and 0.4 M to obtain films of varying thicknesses. This solution was passed through a 0.45  $\mu\text{m}$  PTFE filter prior to spin coating. The spin coated films were annealed at 350 °C in air. To prepare the PEDOT:PSS interfacial layer, a PEDOT:PSS solution (Sigma–Aldrich) was filtered through a 0.45  $\mu\text{m}$  cellulose filter and subsequently spin coated at 5000 r.p.m. for 60 s to obtain 50 nm thick PEDOT:PSS films. The PEDOT:PSS films were then baked at 140 °C for 20 min in the glovebox. A 1:0.8 w/w blend of P3HT (Rieke Metals) and PCBM (Nano-C), dissolved in ortho-dichlorobenzene, was subsequently spin coated onto the NiO or PEDOT:PSS films

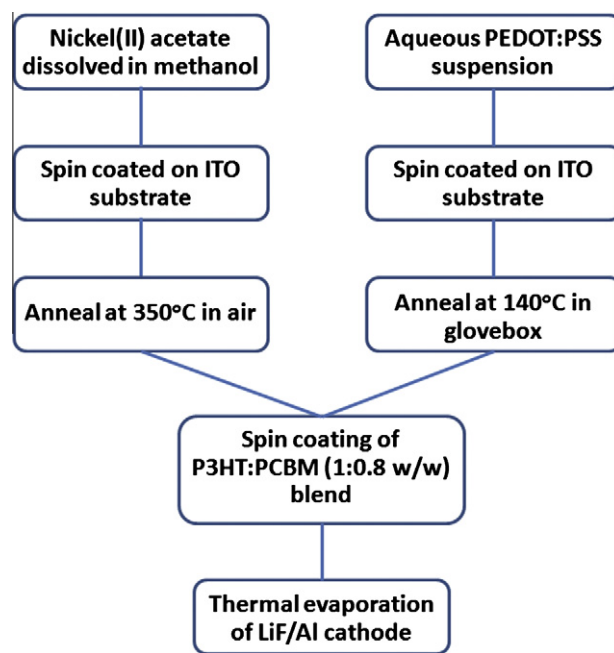


Fig. 1. Schematic work flow for the preparation of NiO (left) and PEDOT:PSS (right) devices.

in the glovebox to obtain a 130 nm photoactive layer. Devices were then completed by sequential thermal evaporation of 0.8 nm LiF and 80–100 nm Al cathode without breaking vacuum ( $\sim 10^{-4}$  Pa). A shadow mask was used during evaporation to make 6 devices of  $3 \times 3 \text{ mm}^2$  each per ITO substrate. A schematic of the experimental work flow is presented in Fig. 1. Device current–voltage measurements were performed in the glovebox with a Keithley 2400 sourcemeter and a solar simulator calibrated to 1 Sun, AM1.5G.

X-ray photoelectron spectroscopy (XPS) measurements were performed with non-monochromatised 1253.6 eV  $\text{MgK}\alpha$  radiation (VG ESCALAB MKII spectrometer). All XPS spectra were calibrated by fixing the C1s spectra (C–C bond) to the reference value of 284.50 eV. Atomic force microscopy (AFM) (Veeco Nanoscope IV), operated in tapping mode, was used to measure film roughness while film thickness was measured by a surface profiler (KLA-Tencor P10). SEM images were obtained with a JEOL JSM-7000F field emission scanning electron microscope (FESEM). Intensity modulated photocurrent spectroscopy was performed in ambient conditions on a controlled-intensity-modulated-photospectroscopy setup (CIMPS) from Zahner Co., Germany. A white LED ( $\lambda_{\text{max}} = 625 \text{ nm}$ ) provided modulated light in the frequency range of 10 Hz–100 kHz superimposed on a steady  $dc$  light intensity of  $10 \text{ W/m}^2$ . Ultraviolet–visible (UV–vis) absorption measurements were performed on a UV–vis spectrometer (Perkin Elmer Lambda 950).

## 3. Results and discussion

Fig. 2 presents the device structure and energy level diagram of P3HT:PCBM devices incorporating the NiO

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