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# Bi-functional lithium doping in dye-sensitized solar cells

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#### Abstract

Despite intensive research into dye sensitized solar cells (DSSCs), simultaneously increasing both the short-circuit photocurrent density ( $J_{sc}$ ) as well as open-circuit photovoltage ( $V_{oc}$ ) of cells has thus far proven impossible. This paper reports the fabrication of Li-doped NiO electrodes to investigate a bi-functional mechanism in which Li enhances light absorption, suspends electron–hole recombination, and reduces the electrical resistivity of the electrode. We also outline the optimization of sol–gel synthesis and investigate the performance of the developed DSSCs. Our results reveal a 6.8% increase in  $V_{oc}$  as well as a 24.7% increase in  $J_{sc}$ , compared with a DSSC with TiO2 electrode. The proposed mechanism, based on state-of-the-art electronic structure theory, provides a valuable guide for the further development of solar cells and electrochemical devices in general.

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#### 1. Introduction

The dye-sensitized solar cell (DSSC) proposed by O'Regan and Grätzel (1991) has numerous advantages over other solar cell technologies, including low production costs, a variety of applicable substrates, and low environmental impact during fabrication [\(Lewis, 2007; Kim](#page--1-0) [et al., 2008; Caramori et al., 2010](#page--1-0)). In principle, the power conversion efficiency  $(\eta)$  of a DSSC is determined by two properties: short-circuit photocurrent density  $(J_{sc})$  and open-circuit photovoltage  $(V_{oc})$ . The former is related to light absorption while the latter is related to electron–hole recombination within the DSSC ([Wang et al., 2004\)](#page--1-0).

High light absorption is desirable as it results in high  $J_{sc}$ . One promising approach to enhancing  $J_{sc}$  involves facilitating the capture of light through methods such as the creation of a light-scattering layer in the working electrode ([Wang et al., 2004; Hore et al., 2006; Ito et al., 2008; Lee](#page--1-0) [et al.,2009; Lan et al., 2011; Yang and Leung, 2011;](#page--1-0) [Chou et al., 2012a; Guo et al., 2012; Liu et al., 2012; Tu](#page--1-0) [et al., 2012; Jeng et al., 2013\)](#page--1-0). To increase the  $V_{oc}$ , two types of energy barrier are commonly applied between the electrode and dye: a wide band semiconductor (such as ZnO) [\(Chou et al., 2012b,c; Guo et al., 2012; Wu](#page--1-0) [et al., 2013\)](#page--1-0) and a semiconductor p-n junction (such as TiO2/NiO) ([Bandara et al., 2005; Chou et al., 2011\)](#page--1-0).

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Another approach involves synthesizing  $TiO<sub>2</sub>$  nanowires with embedded multi-walled carbon nanotubes to increase the transfer of electrons and suppress charge recombination in DSSCs ([Ahn et al., 2013\)](#page--1-0).

Nevertheless, increasing  $V_{oc}$  by introducing an energy barrier or  $p-n$  junction can decrease  $J_{sc}$  due to an increase in electrical resistivity. Few researchers succeeded in enhancing  $V_{oc}$  and  $J_{sc}$  simultaneously. For example, [Chou et al. \(2014\)](#page--1-0) fabricated a DSSC with a light-scattering layer of Ni-doped  $TiO<sub>2</sub>$  particles, which can extend the retention period of light and reduce the recombination of excited electrons in a DSSC, to enhance  $V_{oc}$  and  $J_{sc}$ simultaneously. Although the p-type semiconductor NiO is commonly used to increase  $V_{oc}$  in DSSCs, it is a Mott–Hubbard insulator with very low electrical conductivity of less than  $10^{-13}$  ( $\Omega$  cm)<sup>-1</sup> at room temperature [\(Terakura et al., 1984\)](#page--1-0). [Wu et al. \(2002\)](#page--1-0) claimed that introducing of  $Ni^{2+}$  vacancies and/or doping with monovalent cations (such as  $Li<sup>+</sup>$ ) could produce a considerable increase in conductivity.

The purpose of Li doping in NiO is to increase the hole concentration (major carrier for the p-type) and decrease the electron scattering (or resistance). The decrease in scattering of electrons can facilitate photo electrons to transport from dye to  $TiO<sub>2</sub>$  through tunneling in a DSSC with  $TiO_2/Li_xNi_{1-x}O$  composite particles. Therefore, this study developed a novel hybrid working electrode with  $TiO_2/Li_xNi_{1-x}O$  composite particles in order to enhance  $V_{oc}$  as well as  $J_{sc}$  in DSSCs. TiO<sub>2</sub>/Li<sub>x</sub>Ni<sub>1-x</sub>O composite particles were prepared by dry mixing  $TiO<sub>2</sub>$  particles with Li-doped NiO  $(Li_xNi_{1-x}O)$  powder, which was synthesized using the sol–gel method. We then investigated the effects of  $Li_xNi_{1-x}O$  synthesis conditions and the mass ratio of  $TiO<sub>2</sub>$  to  $Li<sub>x</sub>Ni<sub>1-x</sub>O$  on the performance of DSSCs. Finally, we compared a DSSC equipped with the proposed  $TiO<sub>2</sub>/$  $Li_xNi_{1-x}O$  hybrid electrode against a typical DSSC with  $TiO<sub>2</sub>$  electrode.

#### 2. Experimental details

### 2.1. Preparation and characterization of  $Li_xNi_{I-x}O$  and  $TiO_2/Li_xNi_{1-x}O$

The procedures involved in producing the various structures of  $Li_xNi_{1-x}O$  powder via the sol–gel method are outlined as follows: (1) a solution of 0.5 M nickel (II) nitrate was prepared by mixing 3.635 g nickel (II) nitrate hexahydrate  $[Ni(NO<sub>3</sub>)·6H<sub>2</sub>O]$  (purity of 99.999%) with

der.

25 mL ethanol (purity of 99.5%); (2) a preset mass of lithium nitrate  $(LiNO_3)$  was added to the 0.5 M nickel (II) nitrate solution and this mixture was used to synthesize the  $Li_xNi_{1-x}O$  powder through the sol–gel method. Table 1 lists the test conditions of synthesizing the  $Li_xNi_{1-x}O$  powder with  $x$  representing the ratio between the concentration of Li and that of  $Li + Ni$ . Photographic images of the  $Li_xNi_{1-x}O$  powder were obtained using a digital camera (Panasonic DMC-LZ2) and micrographs were prepared using a scanning electron microscope (SEM) (HITACHI, S-4700).

 $TiO_2/Li_xNi_{1-x}O$  composite particles were prepared using the Mechanofusion system (Hosokawa Micron Corp. AMS-mini). Titanium dioxide (TiO<sub>2</sub>, Uniregion Biotech P-25) particles (average particle size of 21 nm) and the  $Li_xNi_{1-x}O$  powder were loaded into the chamber of the Mechanofusion system with a rotor speed of 6000 rpm for 1 h. The centrifugal force created by the rotating rotor pushes the TiO<sub>2</sub> particles and the  $Li_xNi_{1-x}O$  powder to the inner wall of the rotation chamber. When the  $TiO<sub>2</sub>$  particles and the  $Li_xNi_{1-x}O$  powder pass through the space between the rounded press head and the inner wall of the rotating chamber, high shear-rate particle-to-particle and particle-to-wall collisions force the  $TiO<sub>2</sub>$  particles and the  $Li_xNi_{1-x}O$  powder to adhere to each other and to the inner walls of the rotating chamber. The scraper blade is then used to scrape this powder mixture off and the product is subjected to continuous and repeated cycles of this procedure in the rotation chamber. The detailed mechanism involved in dry mixing was described in our previous works [\(Chou et al., 2009, 2011\)](#page--1-0). [Table 2](#page--1-0) lists the test conditions under which the  $TiO_2/Li_xNi_{1-x}O$  composite particles were prepared. X-ray diffraction (XRD) patterns of  $Li_xNi_{1-x}O$ powder and  $TiO_2/Li_xNi_{1-x}O$  composite particles were obtained using a powder X-ray diffractometer (Shimadzu, XRD-6000).

## 2.2. Preparation and characterization of DSSC with  $TiO<sub>2</sub>$ /  $Li_xNi_{1-x}O$  hybrid electrode

The procedure for fabricating DSSCs with a  $TiO<sub>2</sub>/$  $Li_xNi_{1-x}O$  hybrid electrode is outlined in the following: (1) colloids of  $TiO_2/Li_xNi_{1-x}O$  composite particles were prepared by mixing 2 g composite particles ([Table 2](#page--1-0)) with 8 mL ethanol, 0.8 mL acetylacetone, and 0.1 mL Triton X-100. This mixture was then homogenized in an ultrasonic homogenizer for a given duration; (2) the homogenized colloids of  $TiO_2/Li_xNi_{1-x}O$  were respectively





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