



Efficient dye-sensitized solar cells with surface-modified photoelectrodes

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Abstract

The influence of a mixed overlayer comprising titanium dioxide (TiO₂) and niobium pentoxide (Nb₂O₅), which was coated on the surfaces of TiO₂ nanoporous photoelectrodes, on the performance of dye-sensitized solar cells (DSSCs) was studied. The DSSCs with the TiO₂:Nb₂O₅-coated photoelectrodes showed an increase in short-circuit current (J_{sc}) and open-circuit voltage (V_{oc}), resulting in a power conversion efficiency of 10.37% compared to 7.90% for the reference cell with a TiO₂-coated photoelectrode. The increase in J_{sc} was due to an improvement in the light harvesting and electron collection efficiency. The suppression of the charge recombination between the injected electrons and the I₃⁻ ions was found to increase the V_{oc} value of the device with the TiO₂:Nb₂O₅-coated photoelectrodes.

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1. Introduction

Since the first report on dye-sensitized solar cells (DSSCs) based on a titanium dioxide (TiO₂) semiconductor (O'Regan and Grätzel, 1991), considerable effort has been devoted to the study of DSSCs due to their low cost and high efficiency. Although an impressive power conversion efficiency (PCE) of 12.3% was recently reported by Yella et al. (2011), further improvements in PCE are necessary to successfully commercialize DSSCs. In DSSCs, PCE is achieved by the ultrafast injection of an electron from a photo-excited dye into the conduction band of a semiconductor (TiO₂), the subsequent completion of dye regeneration, and hole transportation to the counter electrode.

Thus, as a media of dye adsorption, electron transport, and electrolyte diffusion, the nanocrystalline-semiconductor photoelectrode (TiO₂) plays a key role in the operation of DSSCs (Grätzel, 2003). The high surface area of nanoporous TiO₂ provides high loading of the nano-molecular dyes, ensuring sufficient light-harvesting efficiency and high electron injection yield. Intensive studies indicate that the conduction band edge (CBE) of TiO₂ determines the driving force for electron injection and also the photovoltage of the device (Cahen et al., 2000; Hagfeldt et al., 2010). On the other hand, the surface and inner trap states of the nanocrystal influence the electron transport in the photoelectrode and the interface recombination between the photoelectrode and the redox electrolyte (Frank et al., 2004; Chien and Hsu, 2013). The pristine TiO₂ films usually generate lower incident photon-to-current conversion efficiency (IPCE) and short-circuit photocurrent (J_{sc}) than

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expected due to the absence of significant band bending in the nano-sized particles (Hagfeldt and Grätzel, 1995), causing a charge recombination between the photo-injected electrons and acceptors, such as the oxidized dye cations and I_3^- ions in the redox electrolyte. To reduce the charge recombination, which would otherwise lead to remarkable electron loss and performance reduction, ultrathin layers of metal oxides (Palomares et al., 2003; Kay and Grätzel, 2002; Bandara et al., 2005; Wu et al., 2013), metal hydroxides (Yum et al., 2006), organic insulators (Kay and Grätzel, 1993; Wang et al., 2003; Zhang et al., 2005), and metal carbonates (Wang et al., 2006; Sharma et al., 2010; Wu et al., 2007) have been applied to the surface of nanoporous TiO_2 electrodes to improve the performance of DSSCs. Among those surface modifiers, titanium (IV) chloride ($TiCl_4$) is one of the most effective materials and is mostly applied to modify nanoporous TiO_2 electrodes (Sommeling et al., 2006; O'Regan et al., 2007; Lee et al., 2012a). This treatment with $TiCl_4$, which is finally converted to TiO_2 by sintering, leads to an increase in J_{sc} as a result of an improvement in electron injection efficiency and a reduction in the recombination rate. Meanwhile, Nb_2O_5 films were fabricated using an electrochemical anodization method, and applied as the photoanodes of DSSCs (Ou et al., 2012; Rani et al., 2013). Particularly noteworthy was the fact that 4 μm -thick nanoporous Nb_2O_5 networks were realized, and the crisscross morphology was possible. Thereby, the DSSC with the nanoporous Nb_2O_5 films showed a significantly higher efficiency compared to that of the device with a TiO_2 nanotubular photoanodes. It has also been reported that a niobium pentoxide (Nb_2O_5) overlayer on a nanoporous TiO_2 electrode can increase the open-circuit voltage (V_{oc}) due to its ability to retard the interfacial recombination rate (Chen et al., 2001; Zaban et al., 2000; Diamant et al., 2004). Thus, TiO_2 and Nb_2O_5 overlayers on a TiO_2 electrode could serve to improve the J_{sc} and V_{oc} , respectively.

In this study, a mixed overlayer (a surface modification layer), composed of both TiO_2 and Nb_2O_5 , was formed on the surface of a nanoporous photoelectrode to further increase the J_{sc} and V_{oc} of DSSCs. In order to obtain the $TiO_2:Nb_2O_5$ -coated photoelectrode, the photoelectrode was treated with a mixed solution of $TiCl_4$ and niobium (V) ethoxide [$Nb(OEt)_5$], followed by sintering. The photovoltaic properties of the DSSC with this photoelectrode was characterized and compared to those of the reference device, which had a TiO_2 -only overlayer.

2. Experimental details

2.1. Materials

Commercial fluorine-doped tin oxide (FTO; sheet resistance $\sim 7 \Omega/\text{square}$) glass (TCO22-7), TiO_2 pastes for the photoelectrode (Ti-nanoxide T/SP) and scattering layer (Ti-nanoxide R/SP), N719 dye (Ruthenizer 535-bisTBA), and an iodide-based electrolyte (AN-50) were purchased

from Solaronix. $TiCl_4$ and $Nb(OEt)_5$ (Sigma–Aldrich) were selected as precursors of the surface modifiers. Chloroplatinic acid ($H_2PtCl_6 \cdot 5.5H_2O$; Kojima Chemicals) was selected as the platinum (Pt) source. All the chemicals were used without any further purification.

2.2. Fabrication of the DSSCs

To prepare the working electrodes, the FTO glasses were cleaned in a detergent solution by sonication for 20 min, and thoroughly rinsed with deionized water and ethanol. The FTO glasses were immersed in a 40 mM $TiCl_4$ solution at 70 °C for 30 min, and washed with deionized water and ethanol. An active TiO_2 layer was formed on the FTO glass via a doctor-blade coating followed by a calcination process at 500 °C for 30 min. A scattering layer was then also deposited onto the TiO_2 layer and sintered to give a 16- μm -thick photoelectrode (TiO_2/FTO). The TiO_2/FTO photoelectrodes were soaked in a mixed solution (40 mM) of $TiCl_4$ and $Nb(OEt)_5$ with various molar ratios of 10:0, 70:30, 50:50, 30:70, and 0:10 at 70 °C for 30 min. This was followed by washing with deionized water and then sintering at 500 °C for 60 min to give the surface-modified photoelectrodes, i.e., $Ti:Nb(10:0)-TiO_2/FTO$, $Ti:Nb(7:3)-TiO_2/FTO$, $Ti:Nb(5:5)-TiO_2/FTO$, $Ti:Nb(3:7)-TiO_2/FTO$, and $Ti:Nb(0:10)-TiO_2/FTO$, respectively. All solutions for surface modification were prepared prior to use. The surface-modified photoelectrodes were separately immersed in a 0.3 mM N719 solution (in acetonitrile/*tert*-butanol, volume ratio of 1/1) for 24 h to obtain working electrodes. To prepare the counter electrode, a drop of 3 mM H_2PtCl_6 solution (in isopropyl alcohol) was placed on the FTO glass, and then it was calcined at 400 °C for 30 min. The working and counter electrodes were sealed together using a 60- μm -thick hot-melt Surlyn spacer under heat. The electrolyte was introduced into the cells through one of the two small holes drilled on the counter electrodes to give the DSSCs with a 25 mm² active area. The resulting DSSCs were used to obtain photovoltaic properties, impedance spectra, IPCE spectra, and dark current.

2.3. Measurements

X-ray photoelectron spectroscopy (XPS) was performed using VG Multilab ESCA 2000 (ThermoVG Scientific) with Al $K\alpha$ radiation. The C 1s photoelectron peak (binding energy at 284.6 eV) was used as the energy reference. Surface morphology of the photoelectrodes was observed by field emission scanning electron microscopy (FE-SEM, HITACHI S-4800). The photovoltaic current–voltage characteristics were determined under 1 Sun illumination (100 mW/cm², AM 1.5), which was verified by an AIST-calibrated Si-solar cell. The UV–Vis absorption spectra were obtained from a Lambda 750 UV–Vis spectrophotometer (Perkin Elmer). The IPCE results were acquired from IPCE G1218a (PV Measurement). Bode plots were

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