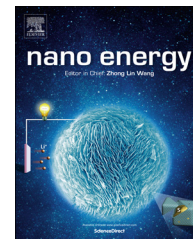


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RAPID COMMUNICATION

# Vanadium oxide as new charge recombination blocking layer for high efficiency dye-sensitized solar cells



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## KEYWORDS

Dye-sensitized solar cells;  
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## Abstract

Vanadium pentoxide ( $V_2O_5$ ) was used as a novel blocking layer in dye-sensitized solar cells (DSCs), leading to a significant efficiency increase from 8.78% to 9.65%. The addition of  $V_2O_5$  layer to nanocrystalline (nc)- $TiO_2$  increased peak external quantum efficiency (EQE) from  $\sim 80\%$  to  $\sim 88\text{--}89\%$ . Cyclic Voltammetry analysis indicated a positive shift of Fermi-level in case of  $TiO_2/V_2O_5$  based cells supported by an increase of its capacitance comparing to bare  $TiO_2$  based cells. Electrochemical impedance spectroscopy (EIS) results exhibited a  $\sim 5$  times higher charge recombination resistance ( $R_{CT}$ ) in  $V_2O_5$  layer modified DSCs than conventional cells, which indicated that back charge transfer from  $TiO_2$  to tri-iodide in the electrolyte was substantially suppressed. Transient photovoltage measurements on conventional and  $V_2O_5$  layer modified cells were conducted and their decays were fitted to calculate the electron recombination lifetime ( $\tau_n$ ), which increased by a factor of  $\sim 3$  in  $V_2O_5$ -based DSCs. This indicated that  $V_2O_5$  significantly reduced the recombination rate at  $TiO_2$ /electrolyte interface, further supporting that  $V_2O_5$  functioned as a new effective surface passivation layer.

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## Introduction

Dye-sensitized solar cells (DSCs) have been attracting great attention since it was invented in 1991 [1–8]. Device

efficiencies beyond 12% have been in reach [9]. In DSCs, the mesoporous  $TiO_2$  is sensitized by a dye, which acts as light absorber [10–14]. The light illuminating on the dye excites electrons from ground to excited states. The excited dye molecules inject electrons into  $TiO_2$  which transport by diffusion before collected by the anode electrode and reach the counter-electrode through an external circuit. Redox

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electrolyte  $I/I_3^-$  is used to reduce the oxidized dye and meanwhile the oxidized redox reduced by taking electrons from the counter-electrode [15].

Power conversion efficiency of DSC device is significantly dependent upon the electron back recombination at  $TiO_2$ /electrolyte interface [16]. Such recombination reduces both the short-circuit current density ( $J_{sc}$ ) and open-circuit voltage ( $V_{oc}$ ), resulting in a decrease in overall power conversion efficiency ( $\eta$ ). Several studies on the electron back recombination were reported recently, [17-20] and some techniques to suppress electron back recombination with the electrolyte have been suggested [16,21-25]. Electron recombination with the electrolyte mainly occurs at two interfaces: FTO substrate/electrolyte and  $TiO_2$  photoanode/electrolyte.

Two main approaches have been used to reduce the back charge transfer from FTO to electrolyte at the FTO/electrolyte interface [22,26]. One is to use a thin  $TiO_2$  compact layer to minimize the exposed FTO surface that is not covered by the nanoporous  $TiO_2$  film. The other approach is to use a blocking material such as poly (phenylene oxide) (PPO) to passivate the FTO substrate using electrodeposition technique [22,26]. At the  $TiO_2$ /electrolyte interface, since the dye molecules need to attach directly to the  $TiO_2$ , the blocking of mesoporous  $TiO_2$  photoanode is basically a little more complicated. Two methods have also been used to decrease the back charge transfer recombination at the  $TiO_2$ /electrolyte interface. The first is to coat the mesoporous  $TiO_2$  film with a thin layer of a wide bandgap semiconductor with more negative conduction band than  $TiO_2$ , while the second is to use a material having an electronic-insulating coating such as  $Nb_2$  and  $CaCO_3$  to form an energy barrier at the  $TiO_2$ /electrolyte interface. This surface barrier reduces the back charge transfer recombination rate and increases device performance [16,23,24,26,27].

In this work, we report a solution-processed and cost-effective vanadium (V) oxide ( $V_2O_5$ ) as a novel charge recombination blocking layer for higher efficiency in DSCs. To the best of our knowledge, this is the first time for  $V_2O_5$  to be used as charge recombination blocking layer. The new type of photoanode consists of mesoporous  $TiO_2$  covered with a layer of  $V_2O_5$ . The conduction band edge of  $V_2O_5$  is significantly more negative than  $TiO_2$  conduction band edge, which indicates that  $V_2O_5$  should function as effective barrier layers for electron recombination at the  $TiO_2$  photoanode/electrolyte interface. As a result, we have found that  $V_{oc}$ ,  $J_{sc}$  and overall efficiency was been significantly improved after applying  $V_2O_5$  to the  $TiO_2$ .

## Experimental section

### Device fabrication

Fluorine doped tin dioxide (FTO) substrates were cleaned using detergent, deionized (DIW) water, acetone, and 2-propanol in an ultrasonic bath for 15 min each. The FTO substrates were treated by submerging in the  $TiCl_4$  aqueous solution (40 mM) for 30 min at 70 °C. Then, a thin compact layer of  $TiO_2$  was spin coated onto the cleaned FTO substrates, followed by the deposition of a nanocrystalline  $TiO_2$  (Ti-Nanoxide HT/SP, Solaronix) via doctor blading. The

film was kept in air for ~10 min and then sintered at 475 °C for ~45 min. A  $TiO_2$  light scattering layer was prepared by doctor blading a mixture of 80% Ti-Nanoxide R/SP (particle size >100 nm) with 20% Ti-Nanoxide HT/SP  $TiO_2$  (particle size 8-10 nm) and sintered as above. And then a 40 mM aqueous solution of  $TiCl_4$  was used to treat the photoanode again. Powder sample of  $V_2O_5$  was purchased from Sigma Aldrich and used as received.  $V_2O_5$  was dissolved in 5 ml acetonitrile to prepare a solution with a  $V_2O_5$ : acetonitrile weight ratio of 1:50. This  $V_2O_5$  solution was then stirred with a magnetic bar on a hot plate at 60 °C for 1 h. After that, it was drop casted onto  $TiO_2$  main layer using micro-pipette and waited for 15 s before spin coated with a spin speed of 4000 rpm for 10 s and sintered at 475 °C. Then the film was cooled down to about 80 °C. The final photoanode was immersed in 0.25 mM N-719 dye solution in 1:1 volume ratio of acetonitrile/valeronitrile for 24 h to attach dye. N719 dye (Ruthenizer 535-bisTBA) was ordered from Solaronix. The dye-attached photoanodes were rinsed by acetonitrile to remove any dye aggregation then dried using nitrogen gas. Both anode electrode and counter electrode were then sealed together using a thermoplastic sealant.

0.03 M  $I_2$ , 0.60 M BMII, 0.10 M  $GuSCN$ , and 0.5 M tert-butylpyridine were dissolved in a mixture of acetonitrile and valeronitrile with 85: 15 volume ratio to prepare the electrolyte used in device fabrication [28]. For comparison, conventional DSCs without use of  $V_2O_5$  were also fabricated as reference devices. The devices were illuminated with a mirror attached on their back under a solar simulator with an AM 1.5 filter at a light intensity of 100  $mW\ cm^{-2}$ . The device area was 0.13  $cm^2$ . The incident photon to electron conversion efficiency (IPCE) was recorded by a Newport M-QE Kit system made of a monochromator and a set of lenses. IPCE data were taken with the pace of 1 point per 5 nm.

### Electrochemical impedance spectroscopy measurement

Electrochemical impedance spectroscopy (EIS) measurements were carried out for both conventional and  $V_2O_5$  treated DSCs in the dark on an Ametek Versastat 3200 potentiostat equipped with frequency analysis module (FDA). AC signal with 10 mV amplitude and 0.01 –  $10^5$  Hz frequency range was used. Forward bias close to open circuit voltage  $\approx 0.8$  V was applied with the photoanode negatively biased and counter electrode positively connected. The experimentally obtained impedance data were fitted by equivalent electrical model using EIS Spectrum Analyzer.

### Cyclic voltammetry measurements

The CV measurements were performed using a solution of 10 mM lithium iodide (LiI) and 0.5 mM iodine ( $I_2$ ) as solutes with 0.1 mM tetrabutylammonium hexafluorophosphate dissolved in acetonitrile as a supporting electrolyte. For comparison, the working electrode was  $TiO_2$  film deposited on FTO glass before and after deposition of  $V_2O_5$ , while a platinum wire worked as a counter-electrode and Ag/AgCl as reference electrode.

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