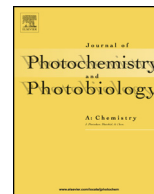




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Electrochemical investigation of Li⁺ ions in the electrolyte on the performance of dyed Mg²⁺-doped TiO₂ solar cells



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ABSTRACT

This work mainly studies the effect of Li⁺ ions in the electrolyte on the enhancement of photoelectric performance and stability in the dyed Mg²⁺ doped TiO₂ solar cells. Measurements of dynamic recombination between interface electron lifetime τ_n and short-circuit current J_{sc} suggested that the doped Mg ions as recombination channels, rather than Li⁺ ions, played key role in recombination reaction in Mg²⁺-doped TiO₂ electrodes. The adsorbed Li⁺ on the surface of Mg²⁺ doped TiO₂ is mainly attributed to the positive shift of the conduction band edge that resulted in the decreased V_{oc} . It is found that the conduction band edge of Mg²⁺ doped TiO₂ electrodes was positively shifted with the increasing concentration of Li⁺ from 0 M to 0.4 M, which led to the increases of the electron injection efficiency η_{inj} and the photocurrent density J_{sc} for Mg²⁺ doped TiO₂ solar cells. And the stability of Mg²⁺ doped in solar cells showed that the adsorbed and intercalated Li⁺ ion in Mg²⁺ doped TiO₂ electrode can improve the cell performance and interface stability.

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1. Introduction

Dye-sensitized solar cells (DSC) is a photoelectrochemical device that converts light into the electrical energy, and the photoelectric conversion efficiency recently obtained over 13%, which exhibits a potential application prospects for its economic and simple preparation process for large-scale productions [1–6]. Under visible light irradiation, the dye molecules become photo-excited and ultrafastly inject electrons into the TiO₂ conduction band. But not all photo-generated electrons are effectively collected, mainly due to the surface broken bonds, surface states and defects in the nanostructured TiO₂ semiconductors, or interface recombination reaction [7]. The nanostructured TiO₂ semiconductors could be modified by aliovalent cation doping and surface treatment to increase the performance of TiO₂/electrolyte interfaces. In recent years, doped TiO₂ with alkaline-earth ions has attracted much attention in the band-edge movement and charge recombination at TiO₂/electrolyte interfaces. Partial pyrolytic deposition of CaCO₃ or wide band gap MgO in nano TiO₂ could

form an effective barrier layer for the core-shell structure, and decrease the recombination reaction for electrons with redox electrolyte to improve the efficiency of solar cells [8,9]. Mg²⁺ ions have relatively small volume, and can be relatively easy to replace Ti⁴⁺ and incorporate into the TiO₂ lattice. The Mg²⁺ doped TiO₂ nanoparticles prepared by hydrothermal method often present higher thermal stability, and obviously negative shift of the conduction band for Mg²⁺ doped TiO₂ resulted in the decreased efficiency of photo-generated electron injection and solar cell performance [10,11].

It is known that cations in the electrolyte can induce the conduction band edge shift and interface property through intercalation into the crystal lattice or chemisorption on the surface of the TiO₂ electrode. Electrolyte cations could modulate the interfacial charge recombination of the nanostructured TiO₂ semiconductor and strongly impacted on the further enhance photovoltaic performance [12,13]. The adsorbed Li⁺ ion on the surface of crystalline intrinsic TiO₂ particles led to the positive shift of the TiO₂ Fermi level and played a role in a shallow trap as a mediator of interfacial charge transfer or recombination center. Recently, we have investigated the doped Mg ions substituted into anatase lattice in dye-sensitized solar cells, and demonstrated that doped Mg ions could enhance the trap states in Mg²⁺-doped TiO₂ as recombination channels [11]. But there is no detailed study about

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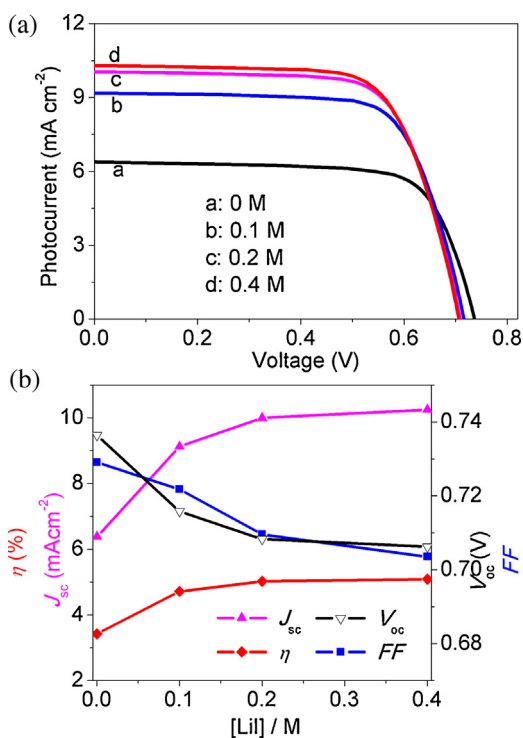


Fig. 1. *J-V* characteristics (a) and photovoltaic performance parameters (b) of the dyed Mg²⁺ doped TiO₂ solar cells with different LiI concentration in the electrolyte.

the effect of Li⁺ in the electrolyte on the Mg²⁺ doped TiO₂ semiconductor interface electrochemical properties and photovoltaic cells performance. In this work, we focused on the effect of Li⁺ ions in the electrolytes on the electron transport and band edge shift for Mg²⁺ doped TiO₂ semiconductor using electrochemical methods. And interface recombination and stability for the dyed Mg²⁺ doped TiO₂ solar cells were also studied.

2. Experimental

2.1 Materials

1-Methylbenzimidazole (MBI) and LiI were obtained from Aldrich. 3-Methoxypropionitrile (MePN) and iodine were purchased from Fluka. Electrolyte compositions were 0.1 M I₂, 0.45 M MBI and 0.6 M DMPII with different concentrations of LiI (ranging from 0 to 0.4 M) in solvent of MePN.

2.1. DSC assembly

Mg²⁺ doped TiO₂ colloidal nanoparticle containing 1 mol% of the doping in this study was prepared by the thermal reaction of titanium tetraisopropoxide and magnesium acetate tetrahydrate in deionized water as described elsewhere [11]. 0.215 g of magnesium acetate tetrahydrate (98%, Sinopharm Chemical Reagent Co., Ltd.) was added in 30 ml of titanium isopropoxide (97%, fluka) under constant stirring. And then the above solution was added dropwise in 50 ml of deionized water under vigorous stirring. A white precipitate was prepared immediately and the formed precipitate solution was kept to form transparent sol. Then, the transparent sol was autoclaved at 200 °C for 12 h to change into gelatin, and the remaining procedures were the same as described in Ref. [11].

Mg²⁺ doped TiO₂ paste was printed using screen printing technique, and sintered in air at 450 °C for 30 min to form Mg²⁺ doped TiO₂ electrodes on FTO glass (TEC-8, LOF). The film thickness was about 12 μm, which was determined by a profilometer (XP-2,

AMBIOS Technology, Inc., USA). After cooling to 80 °C, the thin films were immersed in an acetonitrile solution of dye N719 (0.3 mM) overnight. The excess of N719 dye was rinsed off with anhydrous ethanol before assembly. The counter electrode was platinized by spraying H₂PtCl₆ solution to FTO glass and fired in air at 410 °C for 20 min. Then, it was placed directly on top of Mg²⁺ doped TiO₂ film. The gap between the two electrodes was sealed by thermal adhesive films (Surllyn, Dupont). The electrolyte was filled through the hole in counter electrode, and later the hole was sealed by a cover glass and thermal adhesive films. The total active electrode area of solar cells was about 0.8 cm².

2.2. Methods

The *J-V* characteristics of DSC with the active area of 0.8 cm² were measured under an illumination of AM 1.5 (100 mWcm⁻²) which was realized on a solar simulator (Oriel Sol3A, Newport Stratford Inc., USA, calibrated with standard crystalline silicon solar cell, spectral mismatch was not considered.) with a Keithley 2420 source meter. Intensity-modulated photocurrent/photovoltage spectroscopy (IMPS/IMVS) measurements were carried out on an IM6ex (Germany, Zahner Company) using light-emitting diodes (λ = 610 nm) driven by Expot (Germany, Zahner Company). The LED provided both dc and ac components of the illumination. A small ac component was 10% or less than that of the dc component. The frequency range is 0.1 Hz to 3 kHz. And action spectra of the monochromatic incident photon-to-current conversion efficiency (IPCE) of DSCs were measured with a 300 W xenon lamp (Newport) and a monochromator (Model, Newport 74125, USA) as a function of excitation wavelength over the 350–750 nm spectral range.

3. Results and discussion

3.1. Photovoltaic performance of DSCs

The *J-V* curves of DSC with various amounts of LiI in the electrolytes were shown in Fig. 1 under 100 mWcm⁻² illumination. The addition of LiI in the electrolyte obviously improves J_{sc}, which got a higher photovoltaic performance than that of DSCs without LiI. And photovoltaic performance parameters of the solar cells were not influenced by the further increased LiI content in the electrolytes. The J_{sc} of the solar cells was slightly increased with increasing concentration of LiI from 0.1 M to 0.4 M. And the V_{oc} and FF of the solar cells were decreased with the increase of addition of LiI in the electrolytes. To further elucidate the role of Li⁺, IMPS and IMVS measurements were used to study the interface dynamic mechanism in the dyed Mg²⁺ doped TiO₂ electrodes.

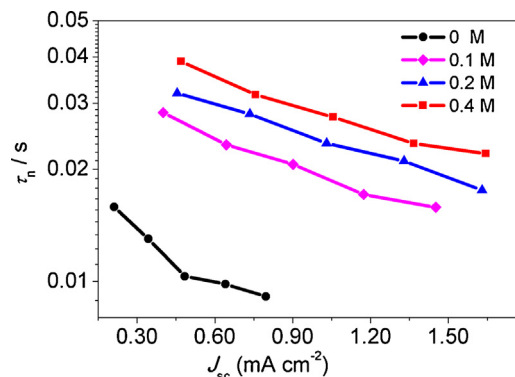


Fig. 2. Electron lifetimes (τ_n) as a function of J_{sc} for DSCs with different LiI concentration in the electrolytes.

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