



Improving the efficiency of CdS quantum dot-sensitized Zn₂SnO₄ solar cells by surface treatment with Al³⁺ ions

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ABSTRACT

The efficiency of CdS quantum dot-sensitized Zn₂SnO₄ solar cells (QDSSCs) was improved by a novel surface treatment with Al³⁺ ions. The driving force for electron injection was strengthened for QDSSCs owing to the reduced conduction band edge of Zn₂SnO₄. The efficiency of Al³⁺ ions-treated solar cells were improved by 44% compared with the untreated ones. Polarization measurements proved the positive movement of the Zn₂SnO₄ conduction band edge. Although the deposited amounts of CdS QDs decreased after the surface treatment, the electron lifetimes were prolonged and the interfacial charge recombination was inhibited, giving rise to a greatly improved short-circuit current and efficiency. By optimizing the number of cycles of chemical bath deposition, a maximum efficiency of 0.263% was achieved under the illumination of one sun (AM1.5, 100 mW cm⁻²).

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1. Introduction

Dye-sensitized solar cells (DSSCs) provide a promising and cost-effective alternative to conventional solar cells because of their high power conversion efficiency, eco-friendly production, easy fabrication and low cost [1]. After the first efficient DSSCs were reported by O'Regan and Grätzel in 1991, many efforts have been made to improve the efficiency of DSSCs, and fundamental research is currently being performed to discover new kinds of DSSCs [2]. Currently, efficiencies exceeding 11% have been obtained by sensitizing nanocrystalline TiO₂ with Ru-based dyes [3,4]. The long-term stability of DSSCs has been greatly improved by utilizing hydrophobic dyes and robust electrolytes [5,6]. Among the new kinds of DSSCs, quantum dot-sensitized solar cells (QDSSCs) have received special attention. For QDSSCs, inorganic semiconductor quantum dots (QDs) absorb light in the visible region and then the excited electrons are injected into the conduction band of TiO₂; thus, the role of the inorganic quantum dots is similar to that of the organic dyes used in DSSCs. Moreover, QDs have additional advantages due to their tunable band structure and absorption spectra [7], their large extinction coefficient [8], and their potential of multiexciton generation [9]. Recently, QDs, such as CdS, CdSe, PbS, and InP, have been widely studied as sensitizers for QDSSCs [10–12]. By co-sensitizing TiO₂ with CdS and CdSe, an efficiency as high as 4.22% was obtained due to the rearranged band structure and increased conduction

band edge [13]. However, even the highest reported efficiencies of QDSSCs are, to date, still far away from highest efficiencies achieved with DSSCs. Hodes compared the various similarities and differences between DSSCs and QDSSCs based on the liquid junction, and he predicted that great improvements in the efficiency of QDSSCs over current values were very feasible and could be expected in the future [14].

Several strategies have been designed to enhance the performances of QDSSCs. For example, in one experiment, a ZnS overlayer was deposited onto CdSe QDs using the chemical bath deposition (CBD) technique to passivate the surface states of CdSe, which resulted in the suppressed charge recombination of photoexcited electrons and holes in the CdSe QDs [15]. In addition, an amorphous TiO₂ overlayer on the QDs improved both the efficiency and stability of QDSSCs [16]. An insulating overlayer of SiO₂ was selectively deposited over the quantum dot-sensitized mesoporous TiO₂ photoanodes and enhanced the efficiency significantly [17]. The introduction of F⁻ ions into the interface of the TiO₂/CdSe QDs provided a straight passage for electron transport, leading to a dramatic elevation of efficiency [18]. However, the photoanodes used in these studies were based on binary oxides. In comparison to simple binary oxides, ternary oxides provide an attractive alternative due to the ease of modifying their chemical properties and band structure. However, studies of ternary oxides as photoanodes are relatively rare. An efficiency of 3.7% was obtained by sensitizing Zn₂SnO₄ photoanodes with a N719 dye [19]. We have previously systematically investigated the performances of Zn₂SnO₄ photoanodes, including the role of co-sensitization with N719 and D131 and the effect of CdS QDs as a sensitizer [20,21]. We have also studied

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the role of Al^{3+} ions in enhancing the efficiency of dye-sensitized Zn_2SnO_4 solar cells [22]. However, it still remains to be observed if surface treatment with Al^{3+} ions is also beneficial for the QDSSCs. In this paper, the influence of treatment of Zn_2SnO_4 photoanodes with Al^{3+} ions on the performance of CdS quantum dot-sensitized Zn_2SnO_4 solar cells is investigated. The decreased conduction band edge of Zn_2SnO_4 after the treatment with Al^{3+} ions was found to favor the electron injection from CdS QDs to the Zn_2SnO_4 conduction band, resulting in a significantly enhanced efficiency. The results presented in this paper provide a new approach for enhancing the performance of QDSSCs.

2. Experimental

All reagents were of analytical grade and were used without further purification. The procedure used to prepare the Zn_2SnO_4 paste can be found in our previous paper [20–23]. The paste was composed of a mixture of nanoparticles that were 20–30 nm in diameter with larger particles that were 50–60 nm in diameter in a molar ratio of 1:1. The paste was printed on FTO glass ($8\text{--}10\ \Omega/\text{sq}^2$, Nippon sheet glass) using a screen printer (Mitani Electronics) and then sintered at $525\ ^\circ\text{C}$ for 2 h. The thickness of the Zn_2SnO_4 film was approximately $5\ \mu\text{m}$. The film thickness was not optimized for performance in this study.

The surface treatment of the Zn_2SnO_4 film with Al^{3+} ions was performed according to our previous report [22]. In a typical procedure, the electrode was immersed in a 0.1 M $\text{Al}(\text{NO}_3)_3$ ethanol solution for 10 min, and then the electrode was rinsed with ethanol three times and sintered at $510\ ^\circ\text{C}$ for 1 h. This procedure was repeated with different loading amounts of Al^{3+} ions. The final electrode is expressed as $\text{Zn}_2\text{SnO}_4/\text{Al}-x$, where x stands for the number of dip-coating-sintering processes used in its fabrication.

The Zn_2SnO_4 photoanodes were sensitized by CdS QDs via chemical bath deposition [21]. The assembly of the cells was described in our previous studies [22]. Namely, the Zn_2SnO_4 electrode was incorporated into a thin-layer, sandwiched solar cell with an active area of $0.25\ \text{cm}^2$. The Pt was sputtered onto FTO glass for use as a counter electrode. A polyethylene spacer, with a thickness of $38\ \mu\text{m}$, was placed between the working electrode and the counter electrode to prevent the cell from short-circuiting. The electrolyte consisted of 0.6 M 1,2-dimethyl-3-n-propylimidazolium iodide, 0.1 M LiI, and 0.05 M I_2 in acetonitrile with 0.5 M 4-tertbutylpyridine. The photovoltaic performance of the solar cells was measured with a source meter (Keithley 2400). A PEC-L11 AM1.5 solar simulator (Pecell, with a 1000 W Xe lamp and an AM1.5 filter) was used as the light source ($100\ \text{mW cm}^{-2}$). Action spectra of the monochromatic incident photo-to-electricity conversion efficiency (IPCE) were collected by a PEC-S20 (Pecell). The incident light intensity was calibrated with a standard for amorphous silicon solar cells that was produced by Japan Quality Assurance Organization.

Electrochemical analysis was performed on an IM6 (Zahner), and the polarization curves of electrodes were measured in 0.1 M Na_2SO_4 electrolyte solution with a scan speed of 5 mV/s. The thickness of the film was determined on a Surfcom 130A (Tokyo seimitsu). The surface chemical states were examined with X-ray photoelectron spectroscopy (XPS, MUTILAB 2000).

3. Results and discussion

The I - V curves of CdS quantum dot-sensitized Zn_2SnO_4 solar cells with and without Al^{3+} ions surface treatments are plotted in Fig. 1, and the corresponding parameters are listed in Table 1. The results indicate that the short circuit current (J_{sc}) increases from 0.49 to $0.64\ \text{mA cm}^{-2}$ (an elevation of 31%), and the power

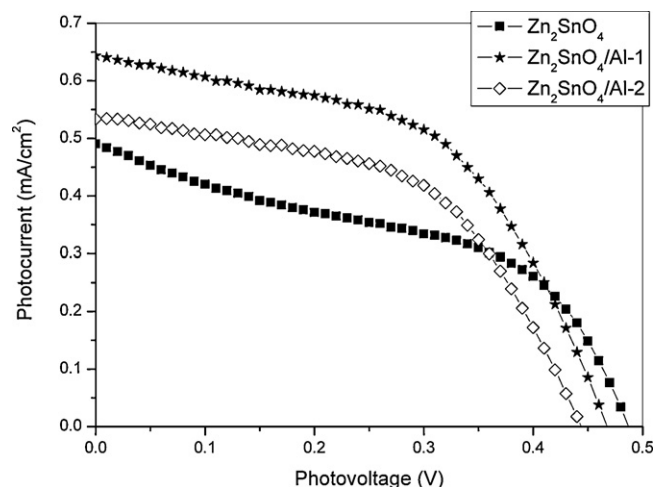


Fig. 1. I - V curves of the solar cells composed of Zn_2SnO_4 with and without the surface treatment of Al^{3+} ions.

conversion efficiency (η) significantly increases by a factor of 44% from 0.109% to 0.157% after the surface treatment with Al^{3+} ions. The observed decrease in the open circuit voltage (V_{oc}) is in agreement with our previous study and is caused by the reduction of a conduction band edge of Zn_2SnO_4 [22]. The variance of the band structure of Zn_2SnO_4 after treatment creates the greatly improved J_{sc} and η due to the enhanced driving force for electron injections. When the Zn_2SnO_4 film was treated twice with Al^{3+} ions, the V_{oc} decreased further. The increase of J_{sc} is more significant than the decrease of V_{oc} , resulting in a notable enhancement of η . In addition, the fill factor (ff) showed a steady increase when the Zn_2SnO_4 film was treated with Al^{3+} ions. The value of ff indicates the quality of the assembled solar cell [24], and a detailed discussion is presented below.

Previously, the decrease of the conduction band edge of Zn_2SnO_4 after the surface treatment with Al^{3+} ions was verified by Mott-Schottky measurements [22]. Herein, we prove the conclusion once more using the polarization test, and the data were collected in a 0.1 M Na_2SO_4 electrolyte solution at a scanning rate of 5 mV/s. When a negative potential was applied to the semiconductor electrode, the charge first fills the blank surface states under the conduction band edge and then accumulates on the space charge of the semiconductor electrode. If the applied negative potential exceeds the flat band edge of the semiconductor, a remarkable increase of cathodic current is observed [25]. Thus, the variance of the conduction band edge, which is followed by the change of the flat band edge, can be determined from the corresponding peaks of cathodic currents in polarization curves. As observed from Fig. 2, the flat band edge of Zn_2SnO_4 and $\text{Zn}_2\text{SnO}_4/\text{Al}-1$ were determined to be -0.329 and $-0.299\ \text{V}$ (vs. Ag/AgCl), respectively. The difference in polarization curves reveals that the flat band edge of Zn_2SnO_4 has a positive shift after the surface treatment of Al^{3+} ions, which is consistent with the results of the Mott-Schottky measurements [22]. The positive shift of the flat band edge of Zn_2SnO_4 demonstrates the decrease of the conduction band edge of Zn_2SnO_4 after the surface treatment, which provides an enhanced driving force

Table 1

Cell parameters of CdS quantum dot-sensitized solar cells composed of Zn_2SnO_4 treated with Al^{3+} for 0–2 times.

Electrode	J_{sc} (mA cm^{-2})	V_{oc} (V)	Fill factor	Efficiency (%)
Zn_2SnO_4	0.49	0.487	0.455	0.109
$\text{Zn}_2\text{SnO}_4/\text{Al}-1$	0.64	0.468	0.521	0.157
$\text{Zn}_2\text{SnO}_4/\text{Al}-2$	0.53	0.444	0.530	0.126

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