

PbS quantum dot solar cells giving high spectral sensitivity in the infrared region

Takaya Kubo¹, Haibin Wang¹, Shouichiro Nakao², Hidenori Saito³, Shinichi Magaino³,
Katsumi Takagi³, Tetsuya Hasegawa², Jotaro Nakazaki⁴, and Hiroshi Segawa^{1,4}

¹The Univ. of Tokyo

4-6-1, Komaba, Meguro-ku, Tokyo 153-8904, Japan

Phone: +81-3-5452-5140 E-mail: ukubo@mail.ecc.u-tokyo.ac.jp

²The Univ. of Tokyo

7-3-1, Hongo, Bunkyo-ku, Tokyo 113-8654, Japan

³Kanagawa Institute of Industrial Science and Technology

705-1, Shimo-Imaizumi, Ebina, Kanagawa 243-0435, Japan

⁴The Univ. of Tokyo

3-8-1, Komaba, Meguro-ku, Tokyo 153-8902, Japan

Abstract

We constructed PbS colloidal quantum dots/ZnO nanowire solar cells that work in the region ranging from the visible to infrared. Highly infrared-transparent conductive oxide (Ta-doped SnO₂; TTO) electrodes were successfully used to enhance solar cells' spectral sensitivity in the infrared region where conventional oxide electrodes such as F-doped SnO₂ and Sn-doped In₂O₃ give strong parasitic absorption. The external quantum efficiency of the solar cells constructed with TTO electrodes reached 50% at the wavelength corresponding to the first exciton absorption peak (1550 nm), which is the highest value ever reported on solution-processed solar cells.

1. Introduction

In recent years, colloidal quantum dots (CQDs) have attracted much attention as light absorbers for next-generation solar cells because mainly of QD-size dependent bandgap tunability, solution process compatibility and so on. Among various types of CQD solar cells, heterojunction CQD solar cells constructed by combining PbS CQDs and ZnO have been widely studied [1, 2], and a power conversion efficiency of 11.23% was reported [2]. In addition to the conventional single-junction solar cell, fundamental studies on CQD solar cells based on a new concept such as multiple exciton generation, hot-carrier concept, and multi-junction concept are carried out [3]. Power conversion efficiency of multi-junction solar cells depends on the number of stacked subcells. A power conversion efficiencies over 50% under one-sun illumination can be achieved by an optimal combination of four or five junctions [4]. The size-dependent bandgap tunability and solution process compatibility are suitable properties to construct low-cost multi-junction solar cells.

Here we try to construct CQD-based bottom subcells for multi-junction solar cells because there are many options for solution-processed top and/or middle subcells such as organic thin film solar cells and perovskite solar cells. PbS QD solar cell performance was also studied by combining sharp-cut filters mimicking middle- and top-subcells as a proof of concept.

2. Experiments

PbS CQDs with different bandgaps were synthesized by following a previous method [5]. The PbS CQD/ZnO NW solar cells were fabricated by using PbS CQDs showing 1550 nm exciton peak by following a previously reported procedure [6]. PbS colloidal quantum dot were infiltrated in the ZnO nanowire layer by a layer-

by-layer method, and an additional QD layer 300 nm in thick was formed by a spin-coating method. Two different transparent conductive oxides (TCOs) were used to fabricate PbS QD/ZnO NW solar cells: one is a widely used F-doped SnO₂ (FTO) electrode, and the other is highly infrared-transparent TCO, Ta-doped SnO₂ transparent oxide (TTO) electrode [7]. TTO thin films grown on glass substrates by pulsed laser deposition using polycrystalline anatase TiO₂ seed layers.

EQE spectra for the wavelength range between 350 nm and 1700 nm were obtained using a spectrophotometer-based machine (Bunko-keiki, CEP-2000MLQ), and fourier transform infrared spectrometer, (JASCO, VIR-300) was employed for the region between 1500 nm and 2000 nm. Current-voltage curves were measured using a solar simulator (Sunko-keiki), which can produce a spectrum matched well with the one-sun spectrum from 350 nm to 2000 nm [6].

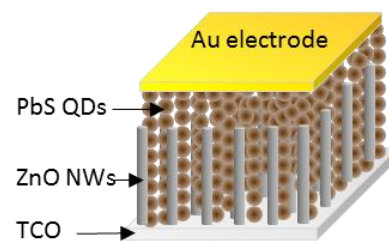
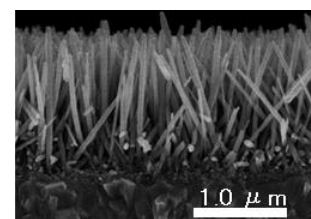


Fig. 1 SEM image of ZnO NWs (upper), and PbS QD/ZnO NW solar cell structure (lower) TCO: FTO or TTO

3. Results and discussion

EQE spectra of the PbS QD/ZnO NW solar cells (FTO-cell, and TTO-cell) give a peak at 1550 nm, originating from the first exciton absorption peak, and show an onset wavelength at 1746 nm (0.71 eV). We employ the wavelength giving 1% EQE as the bandgap wavelength. The EQE of the first exciton peak (1550 nm) obtained on the solar cells with FTO substrates was 20%, whereas the solar cells fabricated with the TTO substrates reached an EQE of 50% that is a record high EQE reported so far on solution-based solar cells. Such a high EQE is chiefly due to high transmittance in the infrared region (Fig. 2).

2%-Ta-doped SnO₂ films give the average sheet resistance of approximately 12 Ω /sq, and the carrier mobility of the film is approximately 70 cm²V⁻¹s⁻¹ higher than the typical mobility of commercially available FTO (ca. 40 cm²V⁻¹s⁻¹). Thanks to the high mobility of TTO layer, the transmittance of TTO is highly transparent not only in the visible region but also in the short-wave infrared region. While the transmittance of FTO, which have been usually used to fabricate solar cells, is attenuated down to 50% at 1.6 μ m (Fig. 2). The TTO-cells then produce higher J_{sc} than the FTO-cells independent of illumination modes (Table 1),

Solar cell performance of the solar cells was also studied under different illumination conditions to examine the potential for the bottom subcell of multi-junction solar cells. In doing so, we focused on the triple-junction solar cells (GaInP/GaAs/InGaAs) that generates a J_{sc} of 14.27 mA/cm² (1-sun) [8], and paid attention to J_{sc} as a measure for current-matching. IV curves of the TTO-cell were measured by illuminating through an 870-nm filter, which mimicks a GaAs middle subcell, to evaluate a potential for the bottom subcell of the triple-junction solar cells. A J_{sc} of 12.6 mA/cm² was obtained, which was approximately 90% of the J_{sc} of the triple-junction solar cells.

Table 1. PV performance obtained under different illumination conditions

filter	w/o (1-sun)		870 nm		1100 nm	
	TTO	FTO	TTO	FTO	TTO	FTO
J_{sc} mA/cm ²	33.1	32.9	12.6	11.3	6.53	5.17
V_{oc} V	0.32	0.32	0.28	0.23	0.25	0.25
FF -	0.44	0.46	0.46	0.47	0.45	0.47
PCE %	4.71	4.85	1.63	1.46	0.75	0.60

* the 870-nm filter and 1100-nm filter mimick a GaAs subcell and Si subcell, respectively.

We also evaluate the TTO-cell performance with using a 1100-nm sharp-cut filter to explore the potential of the bottom cell for 5-junction solar cell [8], wherein the bandgap of each subcell from top to bottom is 2.17 (571)/1.68 (738)/1.40 (886)/1.06 (1169) /0.73 (1698) eV (nm), respectively. J_{sc} of the TTO-cell measured with the 1100-nm filter, mimicking the second subcell from the bottom, is 6.53 mA/cm². This J_{sc} is one third of that of the 5-junction cell. Light absorption in the infrared region has to be enhanced to minimize the difference.

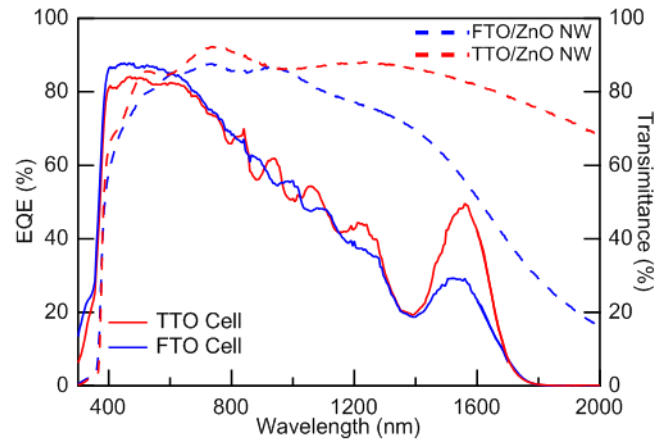


Fig. 2 EQE spectra of TTO- and FTO-cells (left axis), and transmittance spectra of TTO and FTO substrates (right axis).

4. Conclusions

We constructed PbS QD/ZnO NW solar cell using PbS CQDs with the first exciton absorption peak at 1550 nm and a bandgap of 1746 nm (0.71 eV), and succeeded in enhancing EQE in the infrared region by employing Ta-doped SnO₂ electrodes. PV performance of the solar cells with TTO electrodes were measured with different sharp-cut filters to exploit the potential for the bottom subcells of multi-junction solar cells. The results indicates that PbS QD/ZnO NW solar cells including TTO electrodes are promising bottom cells for multi-junction solar cells.

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References

- [1] J. Y. Kim, O. Voznyy, D. Zhitomirsky, E. H. Sargent, *Adv. Mater.*, **25** (2013) 4986.
- [2] M. Liu, O. Voznyy, R. Sabatini, F. P. Garcia de Arquer, R. Munir, A. H. Balawi, X. Lan, F. Fan, G. Walters, A. R. Kirmani, S. Hoogland, F. Laquai, A. Amassian, E. H. Sargent, *Nat. Mater.*, **16** (2017) 258.
- [3] S. Almosnia *et al.*, *Science and Technology of Advanced Materials*, **19** (2018) 336.
- [4] A. Marti, and G. L. Araujo, *Sol. Eng. Mater. & Sol Cells*, **43** (1996) 203.
- [5] H. Wang, T. Kubo, J. Nakazaki, T. Kinoshita, and H. Segawa, *J. Phys. Chem. Lett.*, **4** (2013) 2455.
- [6] H. Wang, T. Kubo, J. Nakazaki, and H. Segawa, *Energy Lett.*, **4** (2017) 2455.
- [7] S. Nakao, N. Yamada, T. Hitosugi, Y. Hirose, T. Shimada, and T. Hasegawa, *Appl. Phys. Express*, **3** (2010) 031102.
- [8] M. A. Green, Y. Hishikawa, E. D. Dunlop, D. H. Levi, J. Hohl-Ebinger, and A. W. Y. Ho-Baillie, *Prog. Photovolt. Res. Appl.*, **26** (2018) 3.