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Effects of Electrolyte on Performance of Quantum Dot-sensitized Solar Cells Using Si Nanoparticles Synthesized by Multi-hollow Discharge Plasma CVD

Si量子ドット増感型太陽電池の電解液最適化

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Effects of electrolyte on performance of Si QDSCs were investigated. We successfully fabricated an efficient polysulfide electrolyte based on the solvent mixed with water and methanol at a volume ratio of 3:7. The optimal electrolyte contains 1.0M Na_2S, 0.5M S and 0.2M KCl. By introducing different ingredients with proper concentration, Jsc, Voc and fill factor are much improved. The efficiency of Si QDSCs using optimized electrolyte is 0.027%.

1. Introduction

Semiconductors show dramatic quantization effects such as tunable band gap and multiple exciton generation when their sizes are below 25nm. Based on these characteristics, Shockley and Queisser limit are expected to be overcome [1]. Therefore, quantum dot-sensitized solar cells (QDSCs) using narrow band gap quantum dots such as Si and CdSe, have attracted considerable interest [2]. Si is a dominant material of solar cells because of its abundance and absence of toxicity. As compared with the crystalline Si solar cells, Si QDSCs have one more advantage of low cost. These backgrounds motivated us to fabricate QDSCs using Si nano-particles [3]. In our previous studies, carrier generation was observed in the wavelength range less than 550 nm in Si QDSCs [4-6]. However the overall performance is still low, especially Voc. Since Voc of the cell is given by the difference between the conduction band edge of TiO_2 layer and the redox potential of the electrolyte, the photovoltaic performance including Voc was much affected by the ingredient and concentration of an electrolyte. Therefore, the performance of Si QDSC was investigated according to various composition of an electrolyte in this study.

2. Experimental

2.1 *Synthesis of Si nano-particles*

Si nano-particles were synthesized by multi-hollow discharge plasma chemical vapor deposition [3]. Hydrogen diluted silane gas was introduced from the bottom of the reactor. It flows through the hollows of the electrodes and pumped out from the top of the reactor. Gas flow rates of H_2 and SiH_4 were 9 and 1.5sccm, respectively. Total pressure was 798 Pa. High voltage with 60MHz was supplied to the electrode. The discharge power was 200 W. In such high H_2 dilution discharges, crystalline Si nano-particles were nucleated and grown in the discharges. They were transported to the downstream region by gas flow and collected with stainless mesh grids located at the downstream region. In this experiment, their mean size was 17 nm.

2.2 *Fabrication of Si QDSCs*

Si QDSCs were fabricated as shown in Fig. 1. FTO substrates were used to make the photo and counter electrodes. The photo electrode was composed of two layers: nano-porous TiO_2 layer and Si nano-particle mixed with TiO_2 (Si/TiO_2) layer.

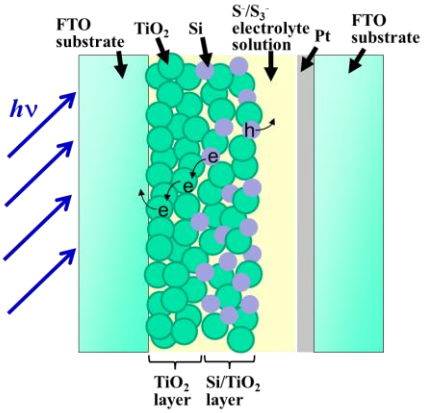

Fig. 1 Structure of Si quantum dot-sensitized Solar cells.

As to an electrolyte, aqueous solvent was replaced by solution mixed with water and methanol at a volume ratio of 3:7. Different ingredients were introduced into this solvent and the donor concentration was also adjusted to find the optimal composition.

3. Results and Discussion

Figure 2 shows I-V characteristics of Si QDSCs according to the concentration of Na_2S. A higher concentration led to a higher Jsc. It was ascribed to the faster hole-recovery with higher S^{2-} concentration. The best efficiency reached to 0.01% with electrolyte containing 1.0M Na_2S.

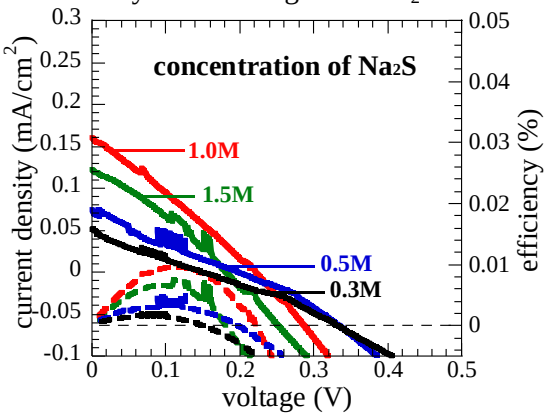

Fig. 2 I-V characteristics of Si QDSCs as a parameter of the concentration of Na_2S.

To enhance the performance of Si QDSCs, other ingredients, S and KCl, were introduced into the electrolyte. I-V characteristics of Si QDSCs with electrolyte containing different concentration of S are shown in Fig. 3 and the characteristic parameters are listed in Table I. Cells with electrolyte containing 1.0M Na_2S and 0.5M S has the best performance. Super-sulfide (S_2^-) has a favorable effect on hole-recovery and the presence of sulfide with S^{2-} leads to the presence of S_2^- [7]. Therefore, a doubled Jsc of 0.36mA/cm^2 is obtained

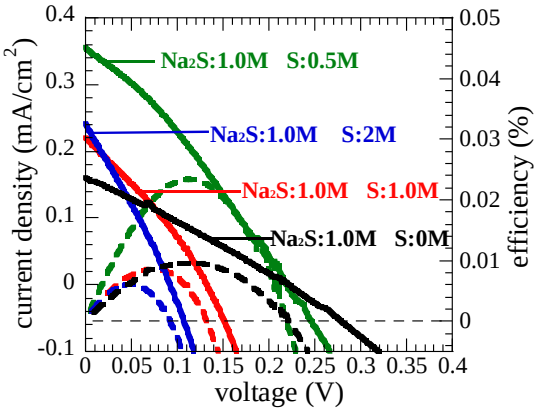

Fig. 3 I-V characteristics of Si QDSCs as a parameter of the concentration of S.

after S is introduced. Fill factor is improved due to the modification of photoelectrode/electrolyte interface, leading to the suppression of charge recombination.

Table I. Characteristics parameters of Si QDSCs with different concentration of S

S (M)	Jsc(mA/cm^2)	Voc(V)	FF	Eff.(%)
0	0.16079	0.222	0.27	0.0096
0. 5	**0.35611**	**0.219**	**0.30**	**0.0234**
1.0	0.22081	0.132	0.29	0.0086
2.0	0.24099	0.092	0.27	0.0060

Since Voc decreases with increasing concentration of Na_2S and S, another ingredient KCl was introduced. The introduction of KCl only contributes to the enhancement of Voc, while the Jsc and fill factor barely has any significant change. The highest performance is achieved with optimized electrolyte containing 1.0M Na_2S, 0.5M S and 0.2M KCl.

Table II. Characteristics parameters of Si QDSCs with different concentration of KCl

KCl (M)	Jsc(mA/cm^2)	Voc(V)	FF	Eff.(%)
0	0.35611	0.219	0.30	0.0234
0.05	0.14453	0.191	0.28	0.0079
0.1	0.32953	0.198	0.30	0.0196
0.2	**0.34513**	**0.252**	**0.31**	**0.027**
0.3	0.33518	0.233	0.33	0.025

4. Conclusions

The optimization of electrolyte for higher performance of Si QDSCs was carried out. We successfully fabricated an efficient polysulfide electrolyte based on the solvent solution mixed with water and methanol at a volume ratio of 3:7. The optimal electrolyte contains 1.0M Na_2S, 0.5M S and 0.2M KCl. By introducing different ingredients with proper concentration, Jsc, Voc and fill factor are much improved. As a result, the efficiency of Si QDSCs using optimized electrolyte is 0.027%.

Acknowledgment

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