Extended Abstracts of the 2010 International Conference on Solid State Devices and Materials, Tokyo, 2010, pp914-915

Development of Multi-Scale Simulation Method for Dye-Sensitized Solar Cells Including Effect of Photoelectrode Material Interface

Mari Onodera[1], Ryo Nagumo[2], Ryuji Miura[1], Ai Suzuki[2], Hideyuki Tsuboi[3], Nozomu Hatakeyama[3], Akira Endou[3], Hiromitsu Takaba[1], Momoji Kubo[4], and Akira Miyamoto[2,1,3]

[1]Tohoku Univ., Dept. Chem. Eng., Grad. School Eng., 6-6-10-205 Aoba, Aramaki, Aoba-ku, Sendai 980-8579, Japan
Phone: +81-22-795-7237 E-mail: mari@aki.che.tohoku.ac.jp
[2]Tohoku Univ., NICHe, 6-6-10-205 Aoba, Aramaki, Aoba-ku, Sendai 980-8579, Japan
[3]Tohoku Univ., Dept. Appl. Chem., Grad. School Eng., 6-6-10-205 Aoba, Aramaki, Aoba-ku, Sendai 980-8579, Japan
[4]Tohoku Univ., FRI, Grad. School Eng., 6-6-11-701 Aoba, Aramaki, Aoba-ku, Sendai 980-8579, Japan

1. Introduction

Dye-Sensitized Solar Cells (DSSCs) are regarded as next-generation solar cells since O'Regan and Grätzel's report [1]. Features of DSSC are low-cost production, wide variety of designs, independence of installation sites, etc. The maximum conversion efficiency of DSSC is about 11% [1]. Further improvement highly relies on better understanding of the energy conversion mechanisms and precise modeling for design optimization [2].

Recent studies showed that under an open-circuit condition, current flow through the interface between TiO_2 and the transparent conducting oxide (TCO) at the photoelectrode occurred due to the recombination of electrons with oxidized redox mediators (I_3^-) at the TCO/electrode interface [3,4]. Therefore, the effect of the TiO_2/TCO interface on the current density (J) - voltage (V) characteristics has been regarded to be significant. The voltage loss at the TiO_2/TCO interface can be modeling by Schottky barrier model based on the thermionic-emission theory [2].

We have developed a multi-scale DSSC simulator [5,6]. This method includes the effects of the irregular TiO_2 porous structure and the electrolyte. However, the internal resistances in construction materials have not been considered.

In this study, we developed the calculation part for the voltage loss at the TiO_2/TCO interface and implemented it to our DSSC simulator. The effect of Schottky barrier height derived from the TCO material on the *J-V* characteristics was evaluated employing the cis-di(thiocyanato)-bis(2,2'-bipyridyl-4,4'-dicarboxylicacid) -ruthenium(II) (N3 dye) as a photosensitizer [7].

2. Methods

Micro-scale simulation: Estimation of oscillator strength of dye

We calculated the oscillator strength of N3 dye using our tight-binding quantum chemical calculation program, "Colors" [8].

Meso-scale simulation(1): Construction of TiO_2 porous structure model

We constructed the irregular TiO_2 porous structure model using the three-dimensional porous structure simulator, "POCO[2]" [9] (Fig. 1).

Meso-scale simulation(2): Electron Diffusion Simulation

The electron diffusion coefficient in constructed TiO_2 porous model was estimated by performing the electron diffusion simulation [5]. The injected electrons from dyes flow in the TiO_2 network toward TCO following the trapping-detrapping process.

Meso-scale simulation(3): Tortuosity Simulation

After donating electrons to the conduction band of TiO_2, the oxidized dye is reduced by I^- in the electrolyte. To take into account the effect of the electrolyte between TiO_2 particles, we calculated the tortuosity of the TiO_2 porous model by the tortuosity simulator [6].

Macro-scale simulation: J-V Characteristics simulation

Using the results of micro- and meso-scale simulations, *J-V* characteristics of DSSC devices was estimated. In this study, we developed the calculation part for the voltage loss at the TiO_2/TCO interface and implemented it in *J-V* characteristics simulator. Fig. 2 shows the previous [6] and

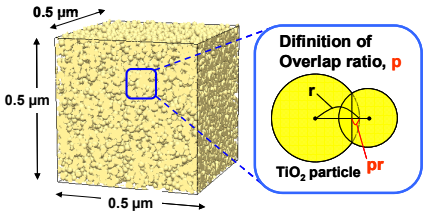

Fig. 1 Meso-scale simulation model.

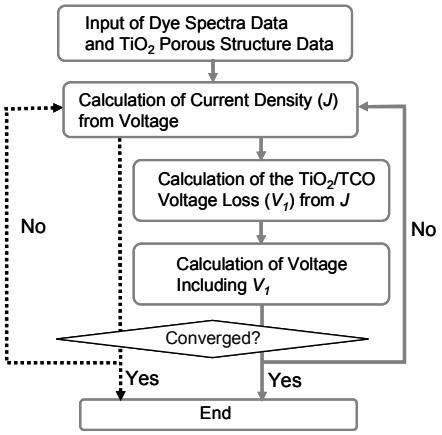

Fig. 2 Simulation flow for *J-V* characteristics. Solid line shows the new method including the effect of TiO_2/TCO interface. Dotted line shows the previous method.

new simulation flow. The voltage loss at the TiO_2/TCO interface V_1 is expressed as Eq. (1) [2],

$$V_1 = \frac{kT}{e}\ln\left[1+\frac{J}{A^*T^2\exp(-e\phi_b/kT)}\right] \quad (1)$$

where k is the Boltzmann constant; T is the absolute temperature; e is the elementary electric charge; J is the current density; A^* is the Richardson constant of TiO_2; Φ_b is the Schottky barrier height.

3. Results and Discussion

We calculated the oscillator strength of N3 dye. This result is same to the previous study and the details have been reported in our previous paper [6].

Meso-scale simulations were performed to the constructed model (Fig. 1). Results and parameters are shown in Table 1.

Using the obtained micro- and meso-results, we simulated *J-V* characteristics. We used the value of 1.3×10^{-10} mol cm^{-2} for the concentration of N3 dye, which was calculated for the monolayer adsorption on an ideal flat surface of TiO_2 [10]. The intensity of incident light $\Phi(\lambda)$ of the AM 1.5 solar spectrum [11] is used for the calculation.

Fig. 3 shows the *J-V* curve comparison between the previous method [6] and the new method including V_1. New method is in better agreement with an experimental result [7] than the previous one. Fig. 4 shows the simulated *J-V* curve with different Φ_b. V_{OC} increases with decreasing Φ_b. Moreover, the effect of Φ_b tends to be significant as Φ_b increases. When Φ_b is 0.5, 0.6, and 0.7 eV, the TCO material is assumed to be Al, FTO, and Au, respectively [12]. In the cases of *J-V* curves corresponding to FTO and Au, simulated *J-V* characteristics are qualitatively consistent with Kron's experimental data [12].

4. Conclusions

In this study, we newly developed a DSSC simulator for *J-V* characteristics including the effect of the TiO_2/TCO interface. Our simulator successfully predicted the effect of Φ_b on V_{OC} as well as *J-V* characteristics. By this improvement, the relationship between the TCO material and the performance of DSSC devices can be shown.

Table 1 Meso-scale simulation results and parameters.

Parameter	Value
Porosity / -	0.5
Overlap ratio / -	0.5
Average radius / nm	20
Number of electrons / -	12,500
Electron diffusion coefficient / $cm^2\ s^{-1}$	1.24×10^{-4}
Tortuosity / -	1.66

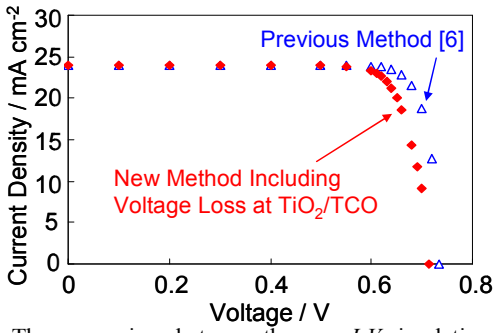

Fig. 3 The comparison between the new *J-V* simulation method including the TiO_2/TCO voltage loss and the previous method.

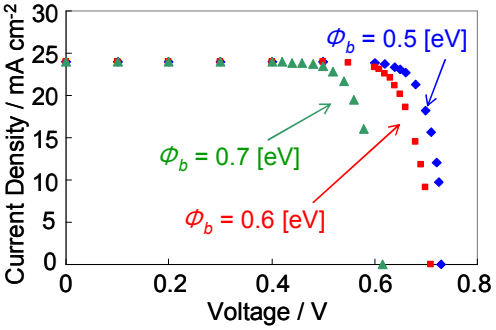

Fig. 4 The effect of TiO_2/TCO Schottky barrier hight on *J-V* characteristics.

References

[1] B. O'Regan, M. Grätzel, Nature **353** (1991) 737.

[2] M. Ni, M. K. H. Leung, D. Y. C. Leung, and K. Sumathy, Sol. Energy Mater. Sol. Cells **90** (2006) 2000.

[3] P. J. Cameron, L. M. Peter, J. Phys. Chem. B **109** (2005) 7392.

[4] P. J. Cameron, L. M. Peter, and S. Hore, J. Phys. Chem. B **109** (2005) 930.

[5] K. Ogiya, C. Lv, A. Suzuki, R. Sahnoun, M. Koyama, H. Tsuboi, N. Hatakeyama, A. Endou, H. Takaba, M. Kubo, C. A. Del Carpio, and A. Miyamoto, Jpn. J. Appl. Phys. **48** (2009) 04C166.

[6] M. Onodera, K. Ogiya, A. Suzuki, H. Tsuboi, N. Hatakeyama, A. Endou, H. Takaba, M. Kubo, and A. Miyamoto, Jpn. J. Appl. Phys. **49** (2010) 04DP10.

[7] M. K. Nazeeruddin, A. Kay, I. Rodicio, R. Humphry-Baker, E. Müller, P. Liska, N. Vlachopoulos, and M. Grätzel, J. Am. Chem. Soc. **115** (1993) 6382.

[8] M. Elanany, P. Selvam, T. Yokosuka, S. Takami, M. Kubo, A. Imamura, and A. Miyamoto, J. Phys. Chem. B **107** (2003) 1518.

[9] M. Koyama, H. Tsuboi, N. Hatakeyama, A. Endou, H. Takaba, M. Kubo, C. A. Del Carpio, and A. Miyamoto, ECS Trans. **7** (2007) 2057.

[10]K. Murakoshi, G. Kano, Y. Wada, S. Yanagida, H. Miyazaki, M. Matsumoto, and S. Murasawa, J. Electroanal. Chem. **396** (1995) 27.

[11]The Simple Model of the Atmospheric Radiative Transfer of Sunshine (online), available from <http://rredc.nrel.gov/solar/spectra/am1.5/>, (accessed 2009-06-15).

[12]G. Kron, U. Rau, and J. H. Werner, J. Phys. Chem. B **107** (2003) 13258.