

Fabrication and Characterization of Perovskite Solar Cells Using Silicon Phthalocyanine Complex [†]

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Abstract: CH₃NH₃PbI₃ perovskite solar cells using silicon phthalocyanine (SiPc) with decaphenylcyclopentasilane (DPPS) were fabricated and characterized. Effects of hole transport capacity of SiPc on the photovoltaic properties were investigated by changing the concentration of the SiPc. Continuous addition of SiPc and DPPS on the perovskite layer improved the short circuit current density related to the conversion efficiency. The conversion efficiencies were stable for 28 days. The photovoltaic performance depended on the (100) crystal orientation and the crystallite sizes.

Keywords: perovskite solar cell; phthalocyanine; photovoltaic properties; morphology

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1. Introduction

To substitute silicon solar cells, perovskite solar cells are expected as the next generation of solar cells, and they are low cost and have easy manufacturing process [1–8]. The perovskite solar cells were constructed with photoactive layer with hole transport layers [9,10]. The photovoltaic characteristics of the perovskite crystals with tuning compositions of chemical elements have been performed to improve conversion efficiencies and microstructures. However, its low stability of the perovskite layer, resulting in reduced power generation characteristics, is a major challenge.

Although 2,2',7,7'-tetrakis(*N,N*-di-*p*-methoxyphenylamine)-9,9'-spirobifluorene (spiro-OMeTAD) is used for the hole transport layer of perovskite solar cells, it is expensive due to its complex synthesis route of multi-step purification process, and it shows low stability against annealing. Alternative hole transport materials using silane derivatives such as decaphenylcyclopentasilane (DPPS) instead of conventional spiro-OMeTAD have been developed for improving stability of the conversion efficiencies while suppressing the decomposition [9–14]. It has also been reported that the self-assembled supramolecules of monoamine porphyrins (MPs, M = Co, Ni, Cu, Zn, and H) have great potential to play a role of improving hole extraction and transport at perovskite grain boundaries, especially for NiP system [15]. In addition, metal phthalocyanines as organic semiconductive materials have advantages to apply the electronic devices such as organic solar cells and perovskite solar cells [16–21]. Addition of the metal phthalocyanines into the perovskite layer will provide promotion of photo-induced carrier generation, charge diffusion related to mobility with suppressing carrier recombination and improving surface morphology of the perovskite layer. Furthermore, basic researches and developments

of the perovskite solar cells using metal phthalocyanines were reported to improve the photovoltaic properties [22–29].

The purpose of this study is to fabricate and characterize the perovskite solar cells using soluble tetrakis(tert-butyl)[bis(trihexylsiloxy) silicon phthalocyanine (SiPc) with decaphenylcyclopentasilane (DPPS) as hole-transporting materials. The effect of SiPc on the photovoltaic properties and formation of perovskite compounds in solar cells were investigated using current density–voltage (J - V) curves under light irradiation, external quantum efficiencies (EQE), X-ray diffraction (XRD) patterns, and optical microscopy images. First-principles molecular orbital calculations were also used for electronic and structural calculations.

2. Materials and Methods

$\text{CH}_3\text{NH}_3\text{PbI}_3$ (MAPbI₃) perovskite solar cells using DPPS and SiPc were fabricated using the following process, as shown in Figure 1. F-doped tin oxide (FTO) substrates were cleaned using an ultrasonic bath with acetone and methanol, and dried under nitrogen gas. The 0.15 and 0.30 M TiO_x precursor solution was prepared from titanium diisopropoxide bis(acetylacetonate) (0.055 and 0.11 mL, Sigma-Aldrich, Tokyo, Japan) with 1-butanol (1 mL, Nacalai Tesque, Kyoto, Japan), and the 0.15 M TiO_x precursor solution was spin-coated on the FTO substrate at 3000 rpm for 30 s and annealed at 125 °C for 5 min. Then, the 0.30 M TiO_x precursor solution was spin-coated on the TiO_x layer at 3000 rpm for 30 s and annealed at 125 °C for 5 min. This process of 0.30 M solution was performed two times, and the FTO substrate was sintered at 550 °C for 30 min to form the compact TiO_2 layer. After that, TiO_2 paste was coated on the substrate by spin-coating at 5000 rpm for 30 s. For the formation of mesoporous TiO_2 layer, the TiO_2 paste was prepared with TiO_2 powder (P-25, Aerosil, Tokyo, Japan) with polyethylene glycol (PEG #20000, Naalai Tesque, Kyoto, Japan) in ultrapure water. The solution was mixed with acetylacetonate (10 μL , Fujifilm Wako Pure Chemical Corporation, Osaka, Japan) and triton X-100 (5 μL , Sigama Aldrich, Tokyo, Japan) for 30 min. The cells were annealed at 120 °C for 5 min and at 550 °C for 30 min to form the mesoporous TiO_2 layer [30,31].

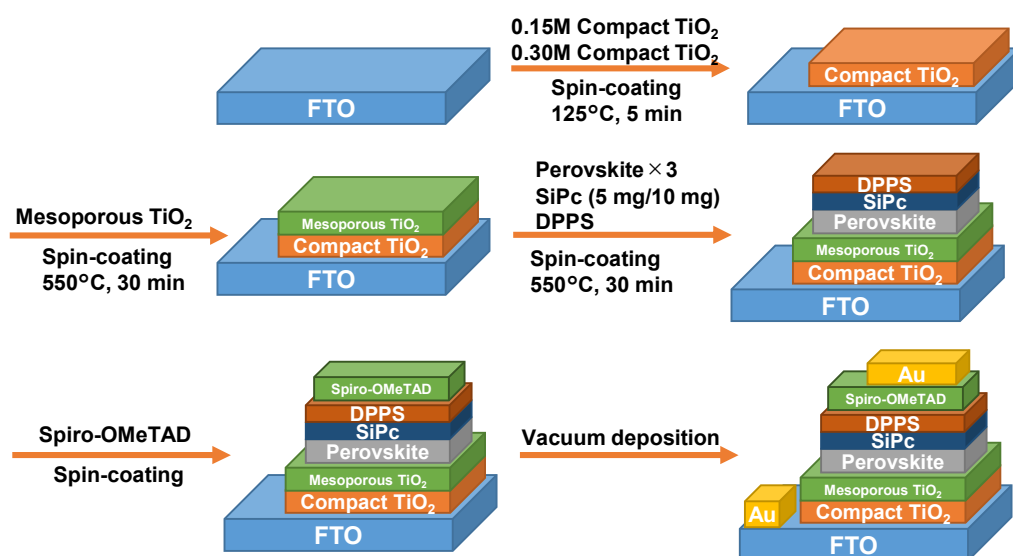


Figure 1. Fabrication process of the perovskite solar cells with SiPc and DPPS.

For the preparation of the perovskite compounds, a solution of $\text{CH}_3\text{NH}_3\text{I}$ (MAI, 2.4 M, Tokyo Chemical Industry, Tokyo, Japan), and PbCl_2 (0.8 M, Sigma Aldrich, Tokyo, Japan) with a desired mole ratio in N, N-dimethylformamide (0.5 mL, Sigma Aldrich, To-

kyo, Japan) was mixed at 60 °C. The solution of perovskite compound was then introduced into the mesoporous TiO₂ by a spin-coating method using an air blowing method [32–34].

SiPc (Orient Chemical Industries, Osaka, Japan) and 2,2',7,7'-tetrakis[N,N-di(methoxyphenyl)amino]-9,9'-spirobifluorene (spiro-OMeTAD, 36.1 mg, Sigma Aldrich, Tokyo, Japan) were used for preparation of the hole transport layer. A solution of silicon phthalocyanine mixed in chlorobenzene (0.5 mL, Fujifilm Wako Pure Chemical Corporation, Osaka, Japan) was prepared by stirring. The DPPS and SiPc solutions were spin-coated by the previously reported method [35,36], and annealed at 190 °C for 5 min.

Additionally, a solution of spiro-OMeTAD (36.1 mg, Sigma Aldrich, Tokyo, Japan) in chlorobenzene (0.5 mL, Fujifilm Wako Pure Chemical Corporation, Osaka, Japan) was mixed with a solution of lithium bis(trifluoromethylsulfonyl)imide (Li-TFSI, 260 mg, Tokyo Chemical Industry, Tokyo, Japan) in acetonitrile (0.5 mL, Sigma Aldrich, Tokyo, Japan) for 30 min. The former solution with 4-tert-butylpyridine (14.4 μL, Sigma Aldrich, Tokyo, Japan) was mixed with the Li-TFSI solution (8.8 μL) for 30 min at 70 °C. All procedures were carried out in ordinary air. Finally, gold (Au) metal contacts were evaporated as top electrodes. Layered structures of the present photovoltaic cells were denoted as FTO/TiO₂/perovskite/SiPc/DPPS/Au.

The *J*–*V* characteristics (Keysight B2901A, Keysight Technologies, Santa Rosa, CA, USA) of the photovoltaic cells were measured under illumination at 100 mW cm⁻² by using an AM 1.5 solar simulator (San-ei Electric XES-301S, Osaka, Japan). The best and average conversion efficiencies, and standard deviations of the solar cells with the three electrodes prepared in this study were measured in the reverse scan of the *J*–*V* curves. The solar cells were illuminated through the side of the FTO substrates, and the illuminated area was 0.080 cm². Microstructural analysis was conducted by an X-ray diffractometer (Bruker, Billerica, MA, USA, D2 PHASER). The surface morphologies of the perovskite layers were examined using an optical microscope (Nikon, Tokyo, Japan, Eclipse E600).

3. Results and Discussion

Figure 2a is a structure model of tetrakis(tert-butyl)[bis(trihexylsiloxy) silicon phthalocyanine. Figure 2b is a structure model after structural optimization by molecular orbital calculation. Energy gaps were also calculated, and their energy levels would contribute the carrier transport [37–39]. SiPc and silicon naphthalocyanine have been used for a *p*-type semiconductor for organic solar cells [40,41].

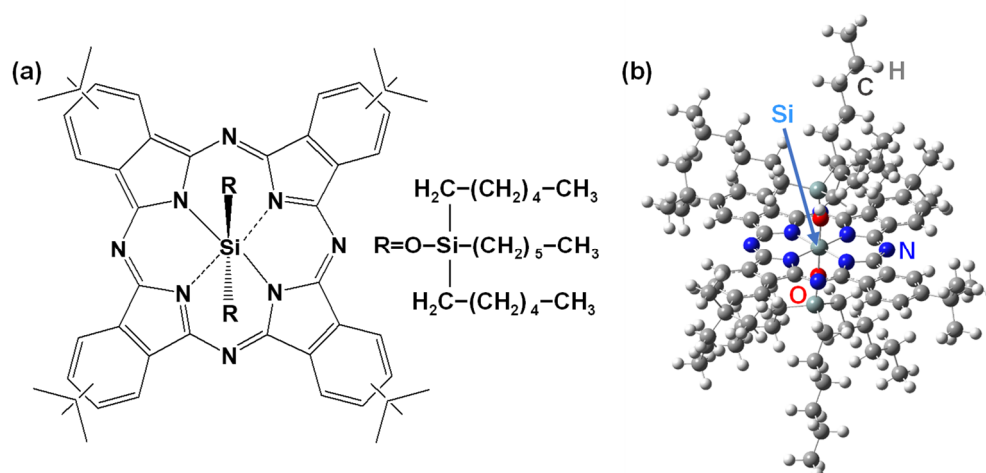


Figure 2. (a) Structure of SiPc. (b) Optimized molecular structure of SiPc.

The *J*–*V* characteristics of the photovoltaic cells were measured. In the case of adding SiPc, the photovoltaic parameters such as short-circuit current density (*J*_{sc}), fill factor (*FF*),

series resistance (R_s), shunt resistance (R_{sh}) and conversion efficiency (η) were obtained to be 20.8 mA cm⁻², 0.819 V, 0.508, 2.97 Ω cm², 127 Ω cm², and 8.66 %. The J - V characteristics of the photovoltaic cells were also measured after 28 days, and the conversion efficiency was stable, which suggests that SiPc layer is related to the stability of the devices.

An X-ray diffraction pattern showed (100) orientation of the perovskite crystals in the devices, and the J_{sc} values would be related with the crystal orientation. Morphologies of the perovskite surface were observed by optical microscopy, and the surface coverage would be affected by the concentration of SiPc solution.

4. Conclusions

Fabrication and characterization of CH₃NH₃PbI₃ perovskite solar cells using SiPc with DPPS were performed. Effects of hole transport of SiPc on the photovoltaic properties were investigated by using the SiPc. Continuous addition of SiPc and DPPS on the perovskite layer improved the short circuit current density related to the conversion efficiency. The stabilities of conversion efficiency were maintained for 28 days. The photovoltaic performance depended on the (100) crystal orientation and the crystallite sizes.

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Conflicts of Interest: The authors declare no conflict of interest.

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