

## Dissertation Abstract

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Dissertation title	<b>Study on Interface and Hole Transport Layer for Efficient c-Si/Organic Heterojunction (HOT) Solar Cells</b> (結晶 Si/導電性高分子接合太陽電池における接合界面およびキャリア輸送に関する研究)		
<p><b>Abstract</b></p> <p>Crystalline silicon (c-Si) based solar cell has reached a power conversion efficiency of 26.33% (latest achievement by Kaneka corporation, Japan), but it requires a high temperature and highly sophisticated processes for the junction fabrication and passivation layer, which involve toxic gases, fabrication complexity and cost. A technologically much less demanding approach, c-Si/Organic heterojunction “HOT” (c-Si heterojunction with organic thin layer) solar cell using solution-processed highly transparent and conductive conjugated polymer poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) (PEDOT:PSS) has been extensively studied compared to the traditional high temperature p-n junction solar cell as it provides a unique possibility to combine high efficiency of crystalline silicon solar cells and low-temperature, solution-processed, technologically less expensive organic photovoltaic devices. The conductivity of this polymer increases up to 1000-1500 S/cm due to the incorporation of various polar solvents e.g. methanol (MeOH) or ethylene glycol (EG) solvent alone, MeOH/EG cosolvents, DMSO and graphene oxide (GO). A power conversion efficiency, <math>\eta</math> of this type of HOT solar cell has reached at 10-13% by adjusting the solvent, the film thickness of PEDOT:PSS, and the resistivity of c-Si substrate without exploiting any light harvesting technique such as textured-Si, AR coating layer, or back-surface field (BSF).</p> <p>However, for further increase in the solar cell performance, several technological schemes should be taken into account. They include efficient light trapping, improved carrier transport property of PEDOT:PSS, reduction of contact resistance of c-Si/cathode interface, back surface field and so on. To solve these matters, the following items have</p>			

been studied in this thesis:

a) The uniform deposition of PEDOT:PSS on textured c-Si: In this work, PEDOT:PSS has been deposited on textured c-Si substrate by chemical mist deposition (CMD) method. The deposition of PEDOT:PSS has been investigated in terms of cavitation frequency  $f$ , solvent, flow rate of nitrogen, substrate temperature, and substrate dc bias as variables for efficient c-Si/PEDOT:PSS heterojunction solar cells. The high-speed-camera and differential mobility analysis characterizations reveals that the average size and flux of PEDOT:PSS mist depend on  $f$ , type of solvent, and substrate dc bias. Film deposition occurs when positive bias is applied to the c-Si substrate at a temperature of 30–40°C, whereas no deposition of films occurs with negative bias, implying that the film is deposited mainly from negatively charged mist. The uniform deposition of PEDOT:PSS films occurs on textured c-Si(100) substrates by adjusting substrate temperature and substrate bias. The adhesion of CMD PEDOT:PSS film to c-Si has greatly been enhanced by applying substrate dc bias compared with that of spin-coated film. The CMD PEDOT:PSS/c-Si heterojunction solar cell devices on textured c-Si(100) exhibits a PCE of 11.0% with better uniformity of the solar cell parameters. Furthermore, efficiency has been increased to 12.5% by the use of isotropically textured-Si and an AR coating layer of MoO<sub>x</sub> formed by CMD. These findings suggest that CMD with negatively charged mist has great potential for the uniform deposition of organic and inorganic materials on textured c-Si substrates by suitably adjusting substrate temperature and substrate bias.

b) The improvement of transport property of PEDOT:PSS by guest molecule: In this topic, the chemistry of amphiphilic perfluorosulfonic copolymer-Nafion coated conductive PEDOT:PSS and its effect on the photovoltaic performance of c-Si/PEDOT:PSS heterojunction solar cells have been demonstrated. The hydrophilic sulfonate group of insulating, chemically stable Nafion interacts with PSS in PEDOT:PSS, which reduce the coulombic interaction between PEDOT and PSS. And the highly hydrophobic fluorocarbon backbone of Nafion favorably interacts with hydrophobic PEDOT of PEDOT:PSS. These factors give rise to the extension of  $\pi$ -conjugation of PEDOT chains. Silver paste used as a top grid electrode diffuses into the Nafion layer and contacted with

underneath Nafion-modified PEDOT:PSS layer. As a consequence, solution-processed Nafion-coated c-Si/PEDOT:PSS heterojunction solar cells exhibit a higher power conversion efficiency of 14.0% with better stability for light soaking rather than that of the pristine c-Si/PEDOT:PSS device by adjusting the layer thickness of Nafion. These findings originate from the chemical stability of hydrophobic fluorocarbon backbone of Nafion, diffusivity of silver paste into Nafion and contact with PEDOT:PSS, and functioning of Nafion as an antireflection layer.

c) The enhanced hole blocking capability at rear Si/cathode interface: In this work, the potential of solution-processed barium hydroxide,  $\text{Ba}(\text{OH})_2$  as a hole blocking layer on the photovoltaic performance of c-Si/PEDOT:PSS front-junction solar cells has been presented. The UPS study shows that  $\text{Ba}(\text{OH})_2$  layer can reduce work function of Al cathode from 4.21 eV to 4.10 eV. The XPS study reveals the thermal stability of the layer. The PCE of the  $2 \times 2 \text{ cm}^2$  size solar cell is increased from 12.8 for pristine to 13.6% using a 2-nm-thick  $\text{Ba}(\text{OH})_2$  layer between c-Si/cathode interface. The PCE is further improved to 14.3% with a short-circuit density  $J_{\text{SC}}$  of  $30.27 \text{ mA/cm}^2$ , an open-circuit voltage  $V_{\text{OC}}$  of 0.632 V, and a fill factor  $FF$  of 0.75 using a  $\text{Ba}(\text{OH})_2$  interlayer between c-Si/cathode and TAPC as a protection layer on top of the device with a remarkable higher stability for air storage compared to the pristine device. This stability improvement originates from the fact that water penetration into the PEDOT:PSS films is effectively blocked by strong ionic interaction between sulfonate anions and protonated bulky TAPC, which in turn, can impede oxidation of this polymer as well as PEDOT:PSS/c-Si interface to a greater extent over time.

d) The efficient back-PEDOT:PSS/c-Si solar cell using  $\text{Ba}(\text{OH})_2$  as HBL and Nafion as passivation layer: In this topic, the promising the back-PEDOT:PSS/c-Si device structure using several techniques has been introduced. The increased PV performance with higher reliability is realizable with the use of back-PEDOT or back-contact device structure to suppress the UV damage to front PEDOT:PSS face and remove the optical loss on top grid electrode. In the device, a ~2-nm-thick spin coated  $\text{Ba}(\text{OH})_2$  has been used as a hole blocking layer at textured Si/Al interface. Nafion has been used as a passivation layer on

front Si surface. The PCE of back-PEDOT:PSS/c-Si solar cells is increased from 4.4% for pristine to 8.1 % with better stability for air storage using Ba(OH)<sub>2</sub> layer in front c-Si/Al interface together with Nafion as a passivation layer. This remarkable increase in PCE mainly originates from the increased  $J_{SC}$  from 23.78 mA/cm<sup>2</sup> to 33.40 mA/cm<sup>2</sup>, and  $V_{OC}$  from 0.536V to 0.573V, which is due to the efficient hole-blocking as well as enhanced electron injection capability of solution-processed Ba(OH)<sub>2</sub> layer and Nafion as an effective passivation layer for silicon. The stability of the back-PEDOT:PSS/c-Si devices is mainly determined by the stability of Nafion passivation over time.