

# Influence on the Haze Effect of Si Thin-Film Solar Cell on Multi-Surface Textures of Periodic Honeycomb Glass

Hyeongsik Park<sup>1,2,3</sup> • Doyoung Kim<sup>1</sup>

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#### Abstract

Double textures were applied to use for multi-junction silicon thin film (Si-TF) solar cells. To improve the longer wavelength, we were formed the microscale periodic honeycomb textured surface on a glass substrate. We achieved the transmittance of 94.26% and found the approximately 52% for haze value. Etched AZO films on the textured glass is crucial role for improving the short-wavelength in Si-TF solar cells. Textured AZO film using a 1% HF solution is suitable for the growth of microcrystalline Si and better light trapping. The transmittance was recorded 80.8% that was 3% higher than that of reference fluorine doped tin oxide (FTO, Asahi-U type) glass. We also achieved for high haze value of 59.8% to enable the light trapping across the entire wavelength region. Using a finite-difference time-domain (FDTD) simulation, we designed a multi-junction Si-TF solar cells with high short-circuit current density of 12.5 mA/cm<sup>2</sup>. To reduce the plasma damage and surface stress on the  $\mu$ c-Si:H layer during growth, we fabricated an a-Si:H/ $\mu$ c-Si:H tandem cell on a multi-surface textures with a  $\mu$ c-Si:H layer of only 1.5  $\mu$ m thick and obtained a conversion efficiency of 13.3%. The developed double textures structure and fabrication technique are expected to improve the performance of various multi-junction Si thin-film solar cells.

Keywords Honeycomb glass · ZnO:Al · Multi-scale · Haze ratio · Tandem solar cells

## 1 Introduction

Light trapping is important for silicon (Si) based solar cells to increase absorption of the photon into the silicon [1–3]. Reducing various loss mechanisms could help to reach the maximum current density ( $J_{SC}$ ) of the solar cells. Yablonovitch and Cody reported the effective enhancement of light intensity by ray optical analysis [4, 5]. Light trapping in Si-based solar cells can lead to an improved blue and red response for the front surface texturing and rear-reflector, respectively. The research in light scattering by etching of zinc oxide (ZnO) goes back to the late nineties (1990s) when

- <sup>2</sup> College of Information and Communication Engineering, Sungkyunkwan University, Suwon 16419, Korea
- <sup>3</sup> Convergence Research Center for Energy and Environmental Sciences, Sungkyunkwan University, Suwon 16419, Korea

Research Center Jülich in Germany and IMT Neuchâtel have competed for the patent and articles. In the meantime, various research group focused on the light trapping in thin film silicon solar cells (TFSSCs) [6–10]. Since then many papers were published on performance of single and tandem junction TFSSCs on textured ZnO or Al-doped ZnO [11–17].

Here, highly efficient Si TF solar cells incorporate a multi-junction structure to reduce thermal relaxation losses and an optical structure to absorb as much incident light as possible. An a-Si:H/µc-Si:H tandem cell consists of a hydrogenated amorphous Si (a-Si:H) top cell for short-wavelength light and a hydrogenated microcrystalline Si (µc-Si:H) bottom cell for long-wavelength light, and is likely the most practical choice for the commercialization of Si TF solar cells [18–20]. In order to improve the solar cell efficiency, optical path length of absorption layer should be maximum and electrical losses should be minimized. Light trapping is the most important feature of Si tandem cells that increases the cell efficiency [21], reduces the absorber layer thickness, and eventually improves the long-term stability of various devices and facilitates low-cost manufacturing. Especially, haze ratio is also essential for the standard of light trapping effect. Haze in the transmittance is the ratio of the diffuse

Hyeongsik Park richspark@skku.edu

Doyoung Kim dykim@uc.ac.kr

<sup>&</sup>lt;sup>1</sup> School of Electrical and Electronic Engineering, Ulsan College, Ulsan 44610, Korea

transmittance to the total transmittance. Earlier approaches are based on the textured surface of the transparent conductive oxide (TCO) [22–24]. The crater-like (fluorine doped tin oxide (FTO, Asahi-U type) and pyramidal texture formation for the front TCO textured surface has a reduction of the haze value at the long wavelength region. Here, the key point is the light trapping effect by a double texture or W-texture (the combination of large and small sized texture), which has been performed the maximum light capturing across the entire wavelength range [25–32].

We previously showed that a glass substrate with periodic honeycomb structures [28, 32, 33] can effectively scatter incident light into rays over a wide range of angles by controlling the surface shape [34]. We demonstrated that the W-texture, nanoscale textures on a TCO layer, and microscale textures consisting of periodic honeycomb patterns on glass substrates can enhance the light absorption in a-Si:H single-junction cells to increase short-circuit current density  $(J_{SC})$ . However, the fabrication of a-Si:H/µc-Si:H tandem cells on a W-texture is yet to be demonstrated. The large curvature of honeycomb glass structures causes surface defects during the chemical texturing of the TCO layer, which in turn leads to crystal defects in the µc-Si:H layer during growth. In accordance with the trade-off relationship between transparency and conductivity, light trapping and high-quality absorption layer growth, plays an important role for solar cell efficiency improvement. In this study, we developed W-texture and a method for depositing a µc-Si:H layer. To minimize the surface defects in the TCO layer and growth defects in the µc-Si:H layer, improvements were made in the chemical treatment of the layer of aluminumdoped zinc oxide (AZO), which is a type of TCO, using a hydrogen fluoride solution, and in the deposition of the bottom µc-Si:H cells using plasma-enhanced chemical vapor deposition (PECVD). Using a finite-difference time-domain (FDTD) simulation, we also designed a current-matching structure for a-Si:H/µc-Si:H tandem cells. Finally, we successfully fabricated the first a-Si:H/µc-Si:H tandem cells on W-texture substrates. The approach of double textured surface structure is to improve the current density of TFSSCs and to reduce the electrical losses.

### 2 Experimental Details

As illustrated in Fig. 1a, an a-Si:H/µc-Si:H solar cell was fabricated on a double textured substrate, where the double texture consisted of microscale periodic honeycomb textured glass and nanoscale textured AZO film. Figure 1b shows the flow chart for a process from etching of glass to the cell fabrication. A glass substrate (Eagle XG, Corning) was sequentially cleaned through sonication using acetone, isopropyl alcohol, and deionized (DI) water, periodic microscale

honeycomb patterns were formed on the glass substrate with a Hexamethyldisilazane (HMDS) and photoresist (PR) hard mask. The glass substrate was placed on spin coater. HMDS solution (2 ml) was deposited and spun at 1000 rpm for 5 s. 2000 rpm for 40 s and 2500 rpm for 5 s, respectively. It was cured for 3 min on a hot plate at the temperature of 245 °C (Real temperature: 150 °C) and then cooled down for 10 min at air ambient. The PR (5 ml) (Model: AZ GXR-601-46 cps) was deposited and spun at 1000 rpm for 5 s, 5000 rpm for 20 s and 2500 rpm for 5 s. The glass substrate with PR was placed in a dry oven and soft baked for 10 min at the temperature of 110 °C. We carried out the pattern formation of PR/HMDS mask layer with Cr Honeycomb mask pattern (The honeycomb pattern:  $3 \mu m \times 3 \mu m$  size, circle type) for UV exposure, that was carried out for 45 s with the pressure of 1.2 kg. In order to transfer the PR/HMDS patterns, developer (Model: AZ 300 Developer) was used by hard baking for 15 min at the temperature of 140 °C.

The 0.5% diluted HF (DI water: HF = 495 ml: 5 ml) etching solution was placed in a double boiler (as shown in Fig. 1c) to maintain the room temperature, 25 °C for the whole chemical reaction. Samples were dipped into the solution and etched at room temperature. After etched glass substrates, we called Etch-1, Etch-2, and Etch-3, which are based on the parameter of etching time as shown in Fig. 1b. SC1 and SC2 cleaning was carried out to remove contaminant on the glass surface after etching [35]. AZO films (Aluminum 1 wt% doped ZnO) were deposited using the radio frequency (RF) magnetron sputtering system for high vacuum chamber. Samples were loaded from load-lock chamber to high vacuum chamber and deposited at the pressure of  $1 \times 10^{-6}$  Torr. For the ZnO:Al seed layer, 100 nm was deposited under the conditions of fixed power of 150 W, Ar gas flow of 20 sccm, working pressure of 12 mTorr, the substrate temperature of 300 °C and the rotation of 5 rpm, respectively. The main AZO film was deposited with the power of 300 W while keeping the other deposition conditions constant. Pre sputtering was performed for 5 min to remove the surface contamination of the sputtering target. A highly transparent and conductive AZO layer (thickness of approximately 630 nm) was then deposited as the TCO layer on the honeycomb textured glass using magnetron sputtering. A double textured surface was prepared by etching the AZO films either by HCl 0.5% or HF 1% in a double boiler to maintain the room temperature for the whole chemical reaction. [28, 36]. Here, we called AZO1 (as deposited AZO), AZO2 (HF 1% etching), AZO3 (HCl 0.5% etching) as a function of different etching solution, respectively.

The a-Si:H top and  $\mu$ c-Si bottom cells were deposited using PECVD on top of the FTO, and the double-texture substrates, respectively. We applied a highly diluted silane-hydrogen (3:150 sccm) mixed gas to deposit the  $\mu$ c-Si:H seed layer at a low growth rate ( $\leq 3$  Å/s). The Fig. 1 a Schematic illustration of the fabricated a-Si:H/  $\mu$ c-Si:H tandem solar cell on the double-texture structure, **b** process flow chart (left side) with an etching shape for the light trapping (right side: SEM images of a cross-section view), and **c** circulating system for etching of glass and ZnO:Al film respectively





(b)



(c)





FTO

700

Wavelength (nm)

(a)

HCl 0.5 % 25 sec etch

HCl 0.5 % 35 sec etch

HCl 0.5 % 45 sec etch

HCl 0.5 % 55 sec etch

800

900

1000

100

80

60

40

20

0

400

500

600

**Fransmittance (%)** 

**Fig. 2** a transmittance and reflectance, with a comparison of the simulation and experiment results, and **b** the transmittance and haze ratio using honeycomb textured glass structures

deposition conditions of the  $\mu$ c-Si:H layer were optimized (high pressure, 3 Torr; low power, 150 W; high frequency, 60 MHz) to minimize the amount of plasma. We also maintained the crystallinity (~44%) of the  $\mu$ c-Si:H layer on the multi surface texture substrates to slightly less than that on conventional FTO glass, such that the proper amount of a-Si:H could passivate the defects at the grain boundary of the crystalline Si and reduce the amount of stress in the crystalline Si during growth. *p*- and *n*-doped  $\mu$ c-Si:H was used for the tunnel junction layers [37]. Finally, the back metal (Ag and Al) layers were deposited using RF magnetron sputtering with a shadow mask to define the cells measuring 5×5 mm<sup>2</sup> in size. The electrical properties such as sheet resistance, resistivity, Hall mobility and carrier

Fig. 3 a Optical transmittance and b haze ratio of AZO films etched by HCl 0.5% as a function of the etching time

(b)

concentration were obtained using 4-point probe system (Model: AIT CMT-100MP) and Hall measurement system (Model: SCINCO HMS 3000). For the optical analysis, Haze meter was used to measure transmittance, parallel transmittance, scattering transmittance and reflectance. We observed the surface status of the TCOs using a field-emission scanning electron microscope (FE-SEM; JSM-7600F, JEOL, and TESCAN LYRA3 and MAIA3). The current–voltage measurement is performed under 100 mW/cm<sup>2</sup> (McScience K3000) AM 1.5 spectrum and the external quantum efficiency is analyzed by the spectral response measurement system (Model: QEX7, PV Measurements Co. Ltd) at the room temperature.

**Fig. 4** FE-SEM images of AZO films etched by HCl 0.5% for **a** 25 s, **b** 35 s, **c** 45 s, and **d** 55 s, respectively



## **3** Result and Discussion

Figure 1b shows the etching structure of Etch-1, Etch-2, and Etch-3 used in our experiment, as well as the transmittance and reflectance as a function of the etching structure as compared with the simulation and experiment results, as shown in Fig. 2a. Some of the etching based structures were simulated using the FDTD. As mentioned above FDTD simulation, which can be different transmittance, reflectance according to the wavelength. The results by using the transverse mode were compared with experimental results as the relevant structure is related to the cross-section of 2-dimensional. As the simulation results indicate, we achieved a transmittance of 90.45%, 89.12%, and 94.26% as a function of Etch-1 through Etch-3, which have 9.5%, 10.49%, and 5.69% reflectance, respectively. We also found similar trends in the transmittance and reflectance as compared with the simulation results, which convincingly demonstrates that our proposal works properly. We analyzed the transmittance and haze ratio of Etch-1 through Etch-3, as shown in Fig. 2b. The transmittance achieved is approximately 94% in the Etch-3 structure, whereas the haze ratio is higher in the Etch-2 structure at approximately 59.69%, and then decreases to approximately 52.1% for the Etch-3 structure. For the high haze ratio of Etch-2, the diffuse transmittance is much higher than the specular transmittance in accordance with the higher aspect ratio arising from the increase in the scattering angle [38]. Figure 2b also shows that the light scattering occurs well on the textured surface with large morphological changes. However, the amount of haze as the scattering efficiency is reduced with the texture size decreased [39–41]. Therefore, we have selected for Etch-3 of an optimal structure, which must be selected considering the surface roughness including the TCO layer deposited on the textured structure and the light scattering within the spectral range that is absorbable in a solar cell. However, it is hard to use Etch-2 structure achieved the highest haze value because the deposition of the silicon layer needs to overcome the deposition issue of the notch sides.

A 630 nm-thick AZO film was deposited and then etched using HCl 0.5% solution to optimize the double-texture structure. Figure 3 shows the transmittance and haze ratio of etched AZO films as a function of etching time. The transmittance was saturated at 84.95% for etching time of 45 s and then decreased for 55 s. The haze ratio also increased to 50.7% for etching time of 45 s and then decreased to 46.5% for 55 s. The sheet resistance of a double-texture structure (etched ZnO:Al on textured glass for honeycomb type) was recorded as 8.55 O/sq for etching time of 45 s that was quite similar to that of FTO reference sample that showed



Fig. 5 a Optical transmittance and b haze ratio of AZO films etched by HF 1% as a function of etching time

sheet resistance of 9.1 O/sq, respectively [28, 29, 32]. Figure 4a–d shows FE-SEM images of a double-texture structure. Regardless of etching time, the inside and edge were not textured uniformly, and the etching shapes of edges were deeper and wider. The deposited AZO films at edges were not dense and over attacked during chemical etching. When AZO films were etched with HCl, it was hard to optimize light trapping due to non-uniform textured surface. It also resulted in low absorption of light with electrical losses. However, as shown in Fig. 4a–d, the AZO surface changed near the notch of the Etch-3 because the HCl solution can easily penetrate the grain boundaries of AZO [42]; the uneven surface deteriorated the performance of the Si TF solar cells fabricated using substrate treated by HCl solution. It can be confirmed by chemical bonding structure of hydrochloric acid (HCl) and reaction mechanism [42].

Figure 5 shows the transmittance and haze ratio of a double-texture structure with HF 1% for various etching times. To obtain comparative optical transmittance and sheet resistance to that of reference FTO sample, it was found that the thickness of AZO films should be around 630 nm. ZnO:Al films etched by HF 1% for 10 s, the average transmittance was recorded as 80.8% that was 3% higher than that of reference FTO sample. The haze ratio does not decrease rapidly in longer wavelength due to nano-and micro-sized features [26, 28, 29]. Figure 6 shows the optimized FE-SEM images of AZO films textured with HF 1% for 10 s. It was seen that ZnO:Al films etched uniformly along with a honeycomb textured glass. The sheet resistance of 10.51  $\Omega$ /sq was suitable for the application of thin film silicon solar cells. For the sample etched with HF 1% for 50 s, narrow and deep holes were sparsely formed. These holes create defects and not favorable for the thin film silicon solar cell deposition.

The TCO layer can be treated with a solution of 0.5% HCl or 1% HF to create the nanoscale textures. Table 1 lists the types of TCOs and their total transmittance and haze ratio (the ratio of diffused transmittance to the total transmittance). Compared to the commercial fluorine-doped tin oxide (FTO) shown in Fig. 7a, AZO1, the as-deposited AZO on the honeycomb textured glass substrate, has a wider texture (Fig. 7b). AZO2 was etched using HCl 0.5% (for 35 s) as shown in Fig. 7c. A nanoscale texture was formed on AZO3, which is AZO treated with a 1% HF solution for 10 s (as shown in Fig. 7d); HF solution slowly etched the AZO surface, forming small dimples and thus creating a textured surface. Finally, we have developed a double texture structure on AZO using a 1% HF solution; Fig. 7d shows the smoother textured surface of AZO3, which is suitable for the growth of microcrystalline Si in the a-Si/µc-Si multijunction thin film solar cell.

Figure 8a shows the transmittance and haze ratio of the TCOs at different wavelengths, measured using a haze meter (SCINCO,  $\lambda$  of 350–950 nm). Compared with FTO, AZO1, AZO2, and AZO3 showed a higher transmittance and haze ratio over the wavelength range (700–900 nm) that should be absorbed in the bottom cell. AZO1 showed a high haze ratio at short wavelengths (< 600 nm), although the haze ratios do not always represent an improved near-field scattering by the top cells because of the far-field scattering effect [43–45]. Both AZO2 and AZO3 have nanoscale textures on the surface and are expected to possess better near-field scattering



Fig. 6 FE-SEM images of AZO films by etching with HF 1% for 10 s with various magnifications

Sample	450 nm		800 nm		Average	
	$\overline{T_{\mathrm{T}}(\%)}$	H (%)	$\overline{T_{\mathrm{T}}(\%)}$	H (%)	$\overline{T_{\mathrm{T}}(\%)}$	H (%)
FTO	77.2	24.4	80.7	4.3	80.2	15.2
AZO1	80.2	63.4	83.5	47.8	81.1	56.5
AZO2	78.2	54.7	85.2	26.3	80.8	41.4
AZO3	77.6	62.7	84.9	56.4	80.8	59.8

 $T_{\rm T}$ , total transmittance; *H*, haze ratio (diffuse/total transmittance). The transmittance was measured at 450 and 800 nm; the average values were obtained within the 350–900 nm range

properties than AZO1; thus, the use of AZO2 or AZO3 will benefit the absorption enhancement at the top cell.

Table 1Types of transparentconductive oxide layers andtheir optical properties

It is important to reduce the thickness of the lower cell because most of the effort (such as the time and cost of the manufacturing process) required to fabricate a tandem cell has concentrated on the deposition of the  $\mu$ c-Si:H layer [46]. It should be noted that, in our work, the thickness of the  $\mu$ c-Si bottom layer was only ~ 1.5  $\mu$ m because the nanoscale AZO3 textures could improve the absorption of the bottom cell. Using an FDTD simulation, we investigated the currents generated at both the top and bottom cells of the tandem cell fabricated on AZO3 (see Figs. 8). Figure 8b shows that the top and bottom currents changed as functions of the a-Si:H

thickness, suggesting that a matching  $J_{SC}$  of ~ 12.5 mA/cm<sup>2</sup> can be obtained in a top cell of ~ 300 nm and bottom cell of ~ 1.5 µm. Based on the simulation, we fabricated a tandem cell using these dimensions.

Figure 9a shows the measured current–density–voltage (*J*–*V*) characteristics for a tandem cell (with a size of  $5 \times 5 \text{ mm}^2$ ) using a solar simulator (McScience, k201 Solar Simulator LAB2000) with an open-circuit voltage ( $V_{OC}$ ) of 1.46 V,  $J_{SC}$  of ~12.56 mA/cm<sup>2</sup>, fill factor (FF) of 72.32%, and initial efficiency ( $E_{\rm ff}$ ) of 13.3%. It should be noted that a high  $J_{SC}$  and its resulting high  $E_{\rm ff}$  are due to the excellent light trapping [37, 46–48] at the double-texture of AZO3. Table 2 summarizes the initial *J*–*V* characteristics fabricated Fig. 7 SEM images of various transparent conducting oxides: a commercial fluorine-doped tin oxide (FTO) on glass, and b AZO1 (as-deposited AZO film), c AZO2 (0.5%-HCltreated AZO film), and d AZO3 (1%-HF-treated AZO film); all honeycomb-textured glass (Etch-3)





(b)

(d)

in an a-Si:H/ $\mu$ c-Si:H tandem solar cell using different double-texture substrates. This  $J_{SC}$  value was attainable only when the thickness of the bottom cell was above 2  $\mu$ m of the cell on AZO1–AZO3, however, the cell on AZO1 did not increase normally because of its surface defects for the FF. The external quantum efficiency (EQE) of the fabricated cell (AZO3) was also measured using an EQE system (QEX10, PV Measurements, Inc.). Figure 9b shows that the measured EQE agrees well with the simulated values.

## 4 Conclusion

A double-texture structure was created using wet etching with HF-based solutions, which is based on the combination of the honeycomb textured glass substrate and the nanoscale textured AZO film. The honeycomb textured glass showed high average values for transmittance (80.8%) and a haze ratio (59.8%) within the 400-800 nm range. Using an FDTD simulation, we designed the a-Si/µc-Si tandem cell with a high matching  $J_{SC}$  (~12.5 mA/cm<sup>2</sup>) at the top and bottom cells. The development of nano-sized AZO etching is carried out to improve light trapping in the short-wavelength. When AZO film is etched by HCl, the transmittance can be improved by 5% compared to that of FTO. However, since the AZO is grown on the honeycomb textured glass, the edge is etched deeper and wider and can cause defect for solar cells. The etched AZO layer using HF solution is uniform and can be multi-surface structure on HCTEX glass. The sheet resistance, transmittance and haze ratio are 10.5 O/ sq, 82% and 46.2%, respectively. We developed a PECVD process to reduce the amount of plasma damage and surface



Fig. 8 a Transmittance and haze of four samples: FTO glass, asdeposited AZO, HCl 0.5% treated AZO, and HF 1% treated AZO film on honeycomb textured glass, and b FDTD simulation as a function of a-Si layer thicknesses of Si tandem solar cell

Table 2 Summary of initial current density-voltage (J-V) characteristics of a-Si:H/µc-Si:H tandem junction solar cell under AM 1.5

Sample	$V_{\rm OC}\left({\rm V}\right)$	$J_{\rm SC}~({\rm mA/cm^2})$	FF (%)	Efficiency (%)
FTO	1.41	11.5	72.6	11.77
AZO1	1.45	12.3	68.53	12.22
AZO2	1.48	12.32	71.56	13.05
AZO3	1.46	12.56	72.32	13.26

stress during micro-crystal growth of the µc-Si:H layer, and we successfully fabricated an a-Si/µc-Si tandem cell on the MST substrate. The cell showed a high  $E_{\rm ff}$  of 13.3% using a µc-Si bottom cell with a thickness of only 1.5 µm, which indicates excellent light scattering by the MSTs for



Fig. 9 Performance of the fabricated a-Si:H/µc-Si:H tandem solar cell: a measured J-V characteristics on different textured substrates, and b measured (solid line) and simulated (dashed line) EQE curves of the top (blue) and bottom (red) cells

Wavelength (nm)

(b)

600

800

1000

both short and long wavelengths. We expect that these MST structures can be further used for high-efficiency Si TF solar cells with triple or quadruple junctions.

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