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2013 Appl. Phys. Express 6 022301
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Cost-Effective and Self-Textured Gallium-Doped Zinc Oxide Front Contacts for Hydrogenated Amorphous Silicon Thin-Film Solar Cells

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Received December 13, 2012; accepted January 14, 2013; published online January 29, 2013

We adopt a economical and original method to fabricate self-textured gallium-doped zinc oxide (GZO) front contacts for hydrogenated amorphous silicon (a-Si:H) single-junction solar cells. This technique involves an atmospheric-pressure plasma jet (APPJ) and a dc sputtering process. The electro-optical characteristics of the textured GZO films are mainly controlled by the haze of organosilicon underlayers deposited by the APPJ. The films exhibit an average optical transmittance of about 80% and resistivity below $9.91 \times 10^{-4} \, \Omega \, \text{cm}$. Moreover, compared with flat solar cells, textured cells fabricated on moderate GZO front contacts show 7.9 and 10.9% enhancements in conversion efficiency and short-circuit current density, respectively. © 2013 The Japan Society of Applied Physics

Translucent conductive oxides (TCOs) are essential for silicon-based thin-film solar cells, including hydrogenated amorphous silicon (a-Si:H) single-junction and hydrogenated microcrystalline silicon (µc-Si:H) single-junction solar cells, owing to their outstanding optical and electrical properties.¹² However, in order to improve the conversion efficiency and stability of silicon-based solar cells, it is necessary to fabricate well-textured TCO substrates to improve light trapping effects in the Si absorbers. Recently, zinc oxides (ZnOs) doped with group III elements have attracted significant attention due to their extraordinary electro-optical characteristics and material abundance.² Moreover, ZnO films are more stable than tin-doped indium oxides (ITO) or fluorine-doped tin oxides (FTOs) in the hydrogen and silane (SiH₄) plasma discharge. Consequently, they are desirable for the manufacture of microcrystalline silicon (µ-Si) and amorphous silicon (a-Si) solar cells.³⁴ To date, many achievements have been made for textured ZnO substrates. Textured ZnO films achieved by post wet etching are already well known.⁵ Unfortunately, this method creates much waste of raw materials. Self-textured ZnO doped with boron (BZO) has been successfully fabricated by low-pressure chemical vapor deposition (LPCVD) or metal organic chemical vapor deposition (MOCVD).⁶,⁷ Nevertheless, these techniques have drawbacks in terms of expensive equipment, toxic precursors, and film instability.

In this study, we exhibit an original and cost-effective technique to produce textured gallium-doped ZnO (GZO) front contacts for a-Si:H single-junction solar cells. A bilayer structure of organosilicon/GZO is used to fabricate self-textured GZO films on glass substrates. This bilayer structure is achieved by an atmospheric pressure plasma jet (APPJ) and dc magnetron sputtering. The APPJ system does not need expensive vacuum systems and maintenance which are necessary for low pressure plasma deposition systems.⁸⁻¹¹ The surface morphology and electro-optical characteristics of the films can be effectively controlled by the haze of organosilicon underlayers. The performance of superstrate-type a-Si:H solar cells fabricated on the different textured GZO front contacts is examined by current–voltage measurements. Finally, external quantum efficiency (EQE) spectra are obtained to demonstrate that the textured GZO films can significantly enhance light trapping in a-Si:H solar cells.

The process flow of fabricating self-textured GZO films is as follows. Firstly, the organosilicon underlayers are deposited on $10 \times 10 \, \text{cm}^²$ Corning Eagle XG glass substrates by a 20 kHz APPJ system, as shown in Fig. 1. The RF power is 450 W. The temperature of glass substrates is kept at 75 °C by a hot plate. The hexamethyldisilazane (HMDSN) precursors, easily volatilized at room temperature and low toxic,¹²⁻¹⁵ are carried through the inner tube by Ar carrier gas changed from 120 to 180 sccm. Clean dry air (CDA) is the plasma forming gas, carried through the outside tube, and the flow rate is maintained at 40 sccm. The distance between the nozzle and the substrate is 15 mm and the scan speed is 300 mm/s. The 1-µm-thick GZO films are deposited on the organosilicon layers at 100 °C by dc magnetron sputtering. The ceramic target contains ZnO and 3.2 wt% Ga₂O₃. The dc power density is 3 W/cm² and the working pressure is 2 mTorr during the sputtering. Finally, all the as-grown textured GZO films are thermally annealed at 500 °C in high vacuum ($< 1 \times 10^{-6} \, \text{Torr}$) for 5 min to improve the electro-optical properties. Superstrate-type a-Si:H single-junction solar cells are fabricated by plasma-enhanced chemical vapor deposition (PECVD). The thickness of p-type, intrinsic, and n-type silicon layers are 10, 300, and 30 nm, respectively. The surface morphology of organosilicon underlayers and textured GZO films is investigated by atomic force microscopy (AFM) and scanning electron microscopy (SEM).
respectively. Optical properties are measured by a UV–
visible–NIR spectrophotometer (Hitachi 4100). Electrical
characteristics are examined by Hall measurements in the
van der Pauw configuration at RT. Current–voltage measure-
ments for a-Si:H solar cells are conducted with a dual light
solar simulator (Wacom WXS-220S L2) under an AM1.5G
100 mW/cm² illumination. Incident photo-to-current effi-
ciency is measured in the wavelength range of 350–800 nm to
evaluate external quantum efficiency (EQE).

Figure 2 shows SEM images of organosilicon layers
deposited at different Ar flow rates ([Ar]) increased from
200 to around 500 nm with increasing [Ar]. The film thickness increases from around 200 to around 500 nm with increasing [Ar]. Meanwhile, the organosilicon clusters can also be expanded by increasing [Ar]. According to the results of Benedikt et al., the difference in flow rate between carrier gas and plasma forming
gas can induce vortexes and reactive particles inside the
tube. More active organosilicon particles are brought
onto the substrate with higher [Ar], and therefore they merge
into big clusters effectively on the surface. Substantially, the
optical roughness of the organosilicon layers can also
be controlled by [Ar]. The haze value, measured by a UV–
visible–NIR spectrophotometer, at the wavelength of 550 nm
rises significantly from 0.05 to 0.11% with increasing [Ar].
Here, organosilicon clusters provide not only light-scattering
centers but also the nucleation sites for GZO deposition.
Figure 3 shows top-view and tilted SEM images of 1-μm
thick GZO films deposited on different organosilicon layers.
The island-like shapes on textured GZO films expand
significantly with increasing roughness of organosilicon
underlayers. Organosilicon clusters serve as the nucleation
sites during the sputtering, and therefore GZO can grow
rapidly near the clusters and form these specific textures.
According to our observation during the sputtering depo-
tition, the growth rates of all the textured GZO films are about
1.4 times greater than that of the flat ones. Besides, AFM
shows that the RMS roughness of textured GZO films
increases significantly from 29.0 to 54.4 nm with increasing
underlayer haze.

Figure 4 reveals the Hall measurement results of textured
GZO films as a function of the RMS roughness. The carrier
density shows slight variations with increasing RMS rough-
ness of GZO films. We speculate that the moderate surface
roughness of organosilicon layers does not significantly
affect the activation of Ga atoms and the carrier density in
the films. Hall mobility suffers vast effects from underlayer
roughness. In particular, when the haze value exceeds
0.08%, the mobility of GZO drops to 26.0 cm²V⁻¹s⁻¹. This
mobility degradation is attributed to the presence of a lot of
lattice defects in highly textured GZO films. Moreover,
there are many valleys on the textured GZO surfaces when
the RMS roughness is higher than 48.6 nm, as shown in
Fig. 3, which also significantly damage Hall mobility. The
resistivity of the textured GZO films increases from 8.14 ×
10⁻⁴ to 9.91 × 10⁻⁴Ωcm accompanying the decreasing
trend of Hall mobility. Figure 5(a) shows the optical trans-
mittance and reflectance spectra of textured GZO films.
Most of the films reveal an average transmittance of about
80% in a wide wavelength range. Especially in the near-
infrared region, because that these GZO films have the
moderate carrier concentration, lower than 10²¹ cm⁻³, which
can prevent the strong absorption from free carriers. By
observing the trend of transmittance and reflectance spectra,
the absorption in the visible wavelength range becomes
more obvious with increasing the RMS roughness. This
defect-related absorption results from the optically-assisted
electron transition from valance band to trap states in the
forbidden gap, and it overwhelms the antireflection effects.
from organosilicon and textured GZO films.\textsuperscript{19} As shown in Fig. 5(b), the haze value of textured GZO films in a wide wavelength range can be enhanced effectively by increasing the roughness of organosilicon underlayers. In this study, the height haze value of 32\% at the wavelength of 550 nm is achieved. Meanwhile, the optical roughness can eliminate the interference phenomenon in the optical transmittance spectra owing to Fabry–Perot resonances within the thick GZO films.

Here, we fabricate superstrate-type a-Si:H single cells on these textured GZO films to evaluate their light trapping capability. The current–voltage characteristics of textured and flat a-Si:H solar cells measured under an AM1.5G 100 mW/cm\textsuperscript{2} illumination are shown in Fig. 6. Compared with the flat solar cells, the textured cells fabricated on the GZO front contacts with RMS roughness of 29.0 nm show a 10.9\% enhancement in short-circuit current density (\(J_{sc}\)) and achieve the conversion efficiency of 7.11\%, as shown in the inserted table of Fig. 6. When GZO film RMS roughness is equal to 54.4 nm, the \(J_{sc}\) can be further enhanced by 18.2\%. However, the open-circuit voltage (\(V_{oc}\)) and fill factor (FF)
suffer significant drops when RMS roughness exceeds 29.0 nm. We speculate that highly rough GZO surfaces can cause shunting paths and leakage currents in a-Si:H. We demonstrate that these textured GZO front contacts can efficiently induce light-trapping effects in a-Si:H single-junction solar cells, especially in the wavelength range above 600 nm. We also demonstrate that a-Si:H solar cells deposited on the well-textured front contacts, with an RMS roughness of 29.0 nm, can achieve the conversion efficiency of 7.11%. In this study, the advantages of the technique for textured TCO substrates are its cost-effectiveness and ease of production, and therefore it has a high potential for application in photovoltaic industrial manufacture.

Acknowledgment This work was supported by the National Science Council of Taiwan under Grant NSC 101-3113-E-009-004.

Fig. 6. Current–voltage characteristics and data of a-Si:H solar cells with various GZO front contacts.

Fig. 7. EQE and reflectance spectra of a-Si:H solar cells with various GZO front contacts.

The enhancement of EQE below 550 nm. For the wavelength above 550 nm, the absorption coefficient of a-Si:H is too low to efficiently absorb the incoming light in the thin active layer so the suppressed reflectance mainly results from light-trapping effects. However, the saturated optical absorption of active layers limits the further enhancement of $P_{oc}$ with increasing GZO RMS roughness. The optical interferences due to Fabry–Perot oscillations in thick a-Si:H layers are also eliminated efficiently by these textured structures.

In summary, the economical and self-textured GZO films are produced successfully by depositing organosilicon underlayers through the APPJ in advance. The organosilicon clusters are created purposely to induce island-like textures on the GZO surfaces. Moreover, these GZO films also reveal outstanding electro-optical characteristics and a high haze value. The textured GZO front contacts can efficiently induce light-trapping effects in a-Si:H single-junction solar cells, especially in the wavelength range above 600 nm. We also demonstrate that a-Si:H solar cells deposited on the well-textured front contacts, with an RMS roughness of 29.0 nm, can achieve the conversion efficiency of 7.11%. In this study, the advantages of the technique for textured TCO substrates are its cost-effectiveness and ease of production, and therefore it has a high potential for application in photovoltaic industrial manufacture.

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