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n-type hydrogenated microcrystalline silicon/indium tin oxide back-reflector interfacial characterization for micromorph tandem solar cell applications

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Back-reflectors (BRs) in hydrogenated amorphous Si (a-Si:H)/hydrogenated micro-crystalline Si (μ c-Si:H) solar cells must not only have sufficient properties to efficiently retrieve the unabsorbed light to where it can be used but must also have lesser back-interfacial states, which are needed for efficient carrier collection. In this study, in order to study the effects of μ c-Si:H(n)/transparent conductive oxide-BRs interfacial properties on the performance of a-Si:H/µc-Si:H tandem solar cells, indium tin oxide (ITO)-BRs films are deposited using different reactive gases (argon (Ar), Ar-hydrogen (H₂), and Ar-helium (He)). A higher recombination of μ c-Si:H(n)/ITO-BRs interfaces causes lower open-circuit voltage (V_{oc}) and fill factor (FF). For ITO deposited in Ar-He reactive gas, the higher recombination at the interface of μ c-Si:H(n)/ITO-BRs may be attributed to the high energetic He ion bombardment; whereas, with the use of Ar-H₂ reactive gas, the higher recombination may be attributed to the blocking carrier collection caused by the higher Schottky barrier height. The highest efficiency of 12.04% (Voc: 1422 mV, short-circuit current density: 11.06 mA/cm², FF: 76.56%) is obtained for ITO-BRs using pure Ar gas. It is worth noting that both the interface damage and the Schottky barrier can impact device performances. © 2015 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4921625]

I. INTRODUCTION

With its potential to reduce material and fabrication costs, thin film silicon solar cell technology is considered to be a promising approach for the large-scale deployment of photovoltaics.¹ Most research on thin film silicon solar cell technology has been carried out on tandem structure, using hydrogenated amorphous Si (a-Si:H) and hydrogenated micro-crystalline Si (μ c-Si:H). To improve short-circuit current density (J_{sc}), light-trapping management is needed, such as a textured front transparent conductive oxide (TCO), a highly reflective intermediate, and/or the employment of back-reflectors (BRs).² The BRs play an important role, as the unabsorbed light needs to be efficiently retrieved to where it can be used.¹ Generally, due to their high reflectance, Aluminum (Al) and/or Silver (Ag) deposited on the silicon layer are used as a back reflective electrode.^{3–5} However, the use of Ag- or Al-BRs could lead to the excitation of surface plasmon polaritons (SPPs), caused by the travelling waves of surface charge density at the metal/dielectric interface.^{5–7} To suppress the loss of SPPs, a dielectric of low refractive index, such as doped zinc oxide (ZnO) and indium tin oxide (ITO), is generally inserted between the silicon and metallic surface.^{5,8–12}

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Importantly, prejudicial cell performances, especially of fill factor (FF) and open-circuit voltage (V_{oc}), could be caused by defects at the μ c-Si:H(n)/TCO-BRs interface and damage at the μ c-Si:H(n) under-layer, as well as by the TCO properties themselves.^{12,13} The deteriorated interface μ c-Si:H(n)/TCO or the μ c-Si:H(n) under-layer itself can increase the recombination in the μ c-Si:H(n) layer, or cause the changes in the potential distribution resulting in a higher recombination rate within the cell.¹² The deposition conditions strongly affect the structure and physical properties of the ITO films, μ c-Si:H(n)/ITO interface, and μ c-Si:H(n) under-layer. When introducing oxygen into the argon (Ar) plasma, the μ c-Si:H(n) layer can be damaged, while there is no indication that severe plasma-related damage limits the electrical cell performance of cells deposited with various total gas pressures.¹² Thus, only a few studies have investigated the back-reflector μ c-Si:H(n)/ITO interface based on altering the reactive gas.¹² This was the issue that motivated the present study.

The presence of hydrogen (H₂) gas with Ar in the sputtering chamber increases the number of oxygen vacancies and hence decreases the resistivity of ITO films.¹⁴ Also, plasma parameters, such as plasma potential, electron density, and electron energy, decrease with the addition of H₂ into the Ar plasma.¹⁵ Lower plasma parameters and better electrical properties may produce less detrimental effects on the μ c-Si:H(n)/ITO interface, and reduced series resistance in the devices, respectively.¹⁶ Conversely, the plasma parameters increase with addition of helium (He) into the Ar plasma, due to the large differences in the ionization potentials, and the ionization cross-section peak values of Ar and He.¹⁷ These higher plasma parameters may have a prejudicial effect on the μ c-Si:H(n)/ITO interface.¹⁶ To investigate the impact of the μ c-Si:H(n)/ITO interfacial states and/or bulk properties of ITO itself on the device performances, the utilization of reactive gas, namely, Ar-H₂, Ar, and Ar-He, could thus be candidates.

In this paper, in order to study the effects of μ c-Si:H(n)/ITO-BRs interfacial properties on the performance of a-Si:H/ μ c-Si:H tandem solar cells, ITO-BRs films were deposited using different reactive gas, pure Ar, Ar-H₂, and Ar-He. The dependence of the dark-current-voltage characteristics on temperature was used to study the μ c-Si:H(n)/ITO-BRs interface. This characteristic is utilized to explain the differences observed in the photovoltaic performance of a-Si:H/ μ c-Si:H tandem solar cells with changing BRs structures.

II. EXPERIMENTAL DETAILS

a-Si:H/ μ c-Si:H tandem solar cells were prepared on 3.2 mm thick float glass substrates by 60 MHz very high frequency plasma enhanced chemical vapor deposition. Schematic of configuration of a-Si:H/ μ c-Si:H tandem solar cells is shown in Figure 1. The a-Si:H(i) layers for the top-cell and μ c-Si:H(i) layers for the bottom-cell in the a-Si:H/ μ c-Si:H tandem solar cells were



FIG. 1. Schematic of the a-Si:H/ μ c-Si:H tandem solar cells with Ag-BRs (a), and ITO-BRs (b).

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350 nm and 1700 nm thick, respectively. Front boron-doped ZnO (BZO) film was prepared, using a metal organic chemical vapor deposition technique. ITO-BRs with thickness of 110 nm were deposited using a direct current-pulsed magnetron sputtering method, with 1.23 W/cm² induced power. A sintered ceramic target (99.999% purity, 6 in. in diameter) with a mixture of 90 wt. % In₂O₃ and 10 wt. % SnO₂ was used as the source material. The gas delivery system was designed to inject the additional gas, such as H₂ and He, in accurate steps of 0.1 sccm. The chamber of the sputtering unit was evacuated to a pressure of 2.0×10^{-4} Pa, before injecting Ar, Ar-H₂, and Ar-He at a pressure of 1.3×10^{-1} Pa. The working pressure was controlled by a throttle valve, in order to sustain equal pressure during ITO film depositions. Metal contacts typically 0.3 μ m thick of Ag/0.2 μ m thick of Al were thermal evaporated either directly onto μ c-Si:H(n)-layers (Fig. 1(a)) or onto ITO-BRs (Fig. 1(b)). In order to distinguish the ITO films deposited at different feeding gas, ITO_Ar, ITO_H₂, and Ar-He, respectively.

Spectroscopic ellipsometry (VASE[®], J.A. Woollam, 240 nm $< \lambda < 1700$ nm) was used to measure the thickness of the ITO layer. In order to determine the electrical and optical properties of the bulk properties of ITO films, ITO layers were also deposited on a Corning Eagle glass substrate. Electrical properties such as resistivity were measured using Hall Effect measurement (Ecopia HMS-3000) at room temperature. The optical properties were measured using a UV-Vis Spectrophotometer (Scinco S-3100). The work-function (Φ) of various ITO layers were calculated by Ultraviolet Photoelectron Spectroscopy data with He I ($h\nu = 21.22 \text{ eV}$) source.

Finally, the a-Si:H/ μ c-Si:H tandem solar cells were characterized by current density–voltage (J-V) measurements (in the temperature range of 298–398 K) under dark and under illuminated conditions (Air Mass 1.5 Global with 100 mW/cm² light intensity at 25 °C). The external quantum efficiency (EQE) of a-Si:H/ μ c-Si:H tandem solar cells was measured using a xenon lamp, a monochromator, and optical filters with differential spectral responses to separate the top and bottom cells.

III. RESULTS AND DISCUSSION

Figure 2(a) shows the performance of the a-Si:H/ μ c-Si:H tandem solar cells having either Ag-BRs or ITO-BRs, in which ITO films were deposited with different sputtered gas admixtures, such as Ar, Ar-H₂, and Ar-He. All devices with incorporated ITO-BRs show better performances than those of Ag-BRs. The results indicate that in order to obtain good cell performances, ITO should be included. Here, a slight change occurs with the use of different ITO layers deposited with different gas admixtures. With the incorporated ITO-BRs layers, the best and worst cases of the a-Si:H/µc-Si:H tandem solar cell performance were the Ar gas admixture and the gas Ar-H₂ admixture, respectively. The best cell shows V_{oc} of 1422 mV, J_{sc} of 11.06 mA/cm², FF of 76.56%, and an efficiency of 12.04%. This value was certified by Korea Testing Certification (see Fig. 2(b)). It can be observed that when the Ar and Ar-H₂ gas admixture was used, the FF values reveal the opposite trend to the electrical bulk-ITO properties, as seen in Figs. 2(a) and 3(a). Hotovy et al. proposed that a resistance of ITO-BRs considerably less than 10^{-4} m Ω , as seen in Fig. 3(a), has a negligible influence on the series resistance, and thus on the FF of the devices.¹² This could suggest that the dominant factor for FF when using ITO-BRs may be the μ c-Si:H(n)/ITO-BRs interfacial properties, rather than the ITO bulk properties. This observation concurs with those of Hegedus et al. and Hotovy et al.^{12,13}

In order to get the better understanding about the relation between the FF and the interfacial properties, the dark J-V characterization for four tandem solar cells at the two selected temperatures is shown in Fig. 3(b). At a measured temperature of 398 K, four devices slightly differed, whereby they show the expected exponential voltage dependence in the forward bias voltage of less than 1 V and show a deviation from the exponential voltage dependence at V > 1 V, due to the current limiting mechanism.¹⁸ At a lower temperature of 298 K, the tandem cell with Ag-BRs shows a significant current limiting mechanism.¹⁸ The dark J-V data were



FIG. 2. (a) Performance of the a-Si:H/ μ c-Si:H tandem solar cell having either Ag-BRs or ITO-BRs, in which ITO films were deposited with different sputtered gas admixture, namely, Ar (ITO_Ar), Ar-H₂ (ITO_H₂), and Ar-He (ITO_He); Here, the lines are guide to the eye. (b) Light-current-voltage characteristic of 12.04% efficiency of an a-Si:H/ μ c-Si:H tandem solar cell with ITO_Ar-BRs (certified by Korea Testing Certification).

also analyzed by plotting dV/dJ vs 1/J (presented in Fig. 3(c)), using the following expression:¹⁸

$$\frac{dV}{dJ} = R_s + \left\{ \frac{nkT}{q} * \left(\frac{1}{J} \right) \right\},\tag{1}$$

where *n*, *k*, *T*, and *q* represent diode ideality factor, Boltzmann's constant, measurement temperature, and electrons charge, respectively. Fig. 3(c) indicates that the significant current limiting mechanism leads to the non-linear behavior of the plotting of dV/dJ vs. 1/J, which results in non-ohmic current limitation for the device without incorporated ITO-BRs. In contrast, all the devices with ITO-BRs have a linear relation, with series resistances, R_s, of 8.52, 11.22, and 12.60 Ω .cm² for Ar, Ar-H₂, and Ar-He gas admixtures, respectively, as shown in Fig. 3(d). Thus, the non-ohmic behavior of devices without the incorporated ITO-BRs (Ag-BRs) resulted in FF of ~10% lower than that of devices incorporated with ITO-BRs.

To explain the reason for the changes of V_{oc} with changing reactive gas, the ideality factor, n, and saturation current density, J_0 , were estimated using the following expression:¹⁸

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FIG. 3. (a) Dependence of resistivity and conductivity in ITO films on the different sputtered gas admixtures. (b) Dark J-V characteristic at 398 K and 298 K for a-Si:H/ μ c-Si:H tandem solar cells having either Ag-BRs, ITO_Ar-BRs, ITO_H₂-BRs, or ITO_He-BRs. (c) dV/dJ vs. 1/J for the same four the a-Si:H/ μ c-Si:H tandem solar cells in Fig. 3(b) at 298 K. (d) Saturation current density (J₀), diode ideality factor (*n*), and series resistance (R_s) for the same four devices in Fig. 3(b). (e) Normalized low kinetic energy cut-off of Ultraviolet Photoemission spectroscopy measured on ITO coated glass substrate, in which ITO was deposited with Ar and Ar-H₂ plasma. (f) Band diagram of the ITO/ μ c-Si(n)/ μ c-Si(i) structure with low and high work-function of ITO films.

$$J(V) = J_o \exp\{V - (J * R)/nkT\}.$$
(2)

The calculated results of *n* and J_0 are presented in Fig. 3(d). A significant decrease of J_0 was observed when the device structure changed from Ag-BRs to ITO-BRs. For ITO-BRs structure, J_0 value slightly varies with the changing of gas admixture. The highest value of *n* is ~3.38 for ITO-BRs with pure Ar plasma, while the lowest value of *n* is ~3.34 for ITO-BRs with Ar-He reactive gas. A lower diode ideality factor may arise from a higher recombination at the interface.^{12,13} Compared to pure Ar plasma, the use of Ar-He plasma results in a decrease of the electron density; hence, the electron temperature (T_e) in the plasma increased due to the large differences in the ionization potentials, and the ionization cross-section peak values of Ar and He.¹⁷ Therefore, the incorporation of He⁺ ions causes greater damage at the μ c-Si(n)/ITO interface due to higher energetic ions bombardment.¹⁹ The greater damage at the μ c-Si(n)/ITO

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interface leads to a higher recombination. Generally, the incorporation of H_2 with Ar plasma could cause a decrease in the flux of Ar^+ and an increase in the flux of ArH^+ .^{15,20} The decreases and increases of these species (Ar^+ and ArH^+) lead to a decrease of T_e , and consequently less interfacial damage and less interface recombination than those of pure Ar plasma.^{15,20} This might seem contradictory to the observation of this study, the lower diode ideality factor, *n*, the higher recombination at the interface and in turn, the lower V_{oc} , and FF of the device using Ar-H₂ reactive gas. Thus, in this case, plasma damage did not impact the device performance.

The $\Phi_{\rm ITO}$ deposited in pure Ar or Ar-H₂ plasma was measured to examine the Schottky barrier height at the back-contact, and it is shown in Fig. 3(e). The $\Phi_{\rm ITO}$ increased when using Ar-H₂ reactive gas during the sputtering process. As shown in Fig. 3(f), the simulation demonstrated that the higher $\Phi_{\rm ITO}$ could cause a higher barrier height for electrons collection and a lower "mirror" for holes reflection,^{21,22} possibly leading to high interface recombination and subsequently to a lower *n* and higher J₀, as shown in Fig. 3(d). Consequently, the highest values of V_{oc} and FF were obtained for the lowest $\Phi_{\rm ITO}$ in case of Ar gas admixture.

The EQE of the device structures fabricated with Ag-BRs and two selected ITO-BRs are depicted in Fig. 4(a). As expected, all bottom cells fabricated with ITO-BRs show higher spectral response at the long wavelength region than the Ag-BRs. This result is also reflected in the improvement in the J_{sc} of the ITO-BRs (Fig. 2(a)). Fig. 4(b) shows a comparison of the integrated absorptance of the ITO layers on glass deposited at different gas admixtures. The



FIG. 4. (a) EQE of the top cell and of the bottom cell of a-Si:H/ μ c-Si:H tandem solar cell with Ag-BRs, ITO_Ar-BRs, and ITO_He-BRs. (b) Relative change of J_{sc} of a-Si:H/ μ c-Si:H solar cells with different BRs structures and integrated absorptance of ITO layers on glass, as a function of sputtering gas admixtures.

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highest and the lowest absorptances of ITO layers were obtained for films deposited with Ar-H₂ and Ar-He reactive gas, respectively. The ITO layer deposited with pure Ar gas shows a moderate absorptance. A clear correlation can be seen between the lower ITO absorptance (Fig. 4(b)) and the higher spectral response for the bottom cells at the long wavelength region (Fig. 4(a)). In this study, the tandem cells are bottom-limited. The higher J_{sc} of the bottom cells is good for current matching; hence, the current of the entire tandem cell is the highest with the use of ITO-BRs with Ar-He reactive gas.

IV. CONCLUSION

In summary, we show the superior performance of the light management of ITO-BRs over the conventional Ag-BRs in a-Si:H/ μ c-Si:H tandem solar cells. The dependence of the darkcurrent-voltage characteristics on the temperature analysis suggested that a critical factor that influences device performances, such as FF and Voc, is the interfacial properties rather than the bulk ITO properties. The use of Ar-He plasma could cause interface damage, resulting in higher interface recombination due to high energetic He^+ ions bombardment. Meanwhile, for the high work-function ITO film deposited by using Ar-H₂ plasma, the higher interface recombination is attributed to the higher Schottky barrier height, causing the blocking of carrier transport at the interfacial μ c-Si:H/ITO-BRs. Meanwhile, a higher J_{sc} was observed for the device using ITO-BRs with the lowest absorptance. An a-Si:H/µc-Si:H tandem solar cell of 12.04% efficiency (Voc: 1422 mV, Jsc: 11.06 mA/cm², and FF: 76.56%) was obtained for the ITO-BRs film deposited with pure Ar plasma. It is worth noting that the Schottky barrier, as well as the interface damage, could impact device performances.

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