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Application of $\text{Si}_x\text{N}_y\text{:H}_z$ (SiN) as index matching layer in a-Si:H thin film solar cells

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The difference in refractive indices of glass substrate and transparent conducting oxide (TCO) electrode causes optical reflection in thin film solar cells, which results in lower absorption of light for devices. An anti-reflection layer between glass and TCO is required to reduce the loss of light due to optical reflection. Silicon nitride ($\text{Si}_x\text{N}_y\text{:H}_z$) films have shown antireflection property. The refractive index of $\text{Si}_x\text{N}_y\text{:H}_z$ films can be engineered by changing the silicon or nitrogen content in the film. Here, we report the optimization of refractive index of $\text{Si}_x\text{N}_y\text{:H}_z$ to achieve a value between refractive index of glass (1.5) and TCO film (2.0). $\text{Si}_x\text{N}_y\text{:H}_z$ films have been deposited in a RF-plasma enhanced chemical vapour deposition system operating at a frequency of 13.56 MHz. The substrate temperature was fixed at 300 °C. Fourier transform infrared analysis has been used to determine the nature of Si-N, N-H, and Si-H bonding in the films. Refractive index of films has been measured using spectroscopic ellipsometer. The optical reflectance and transmission of $\text{Si}_x\text{N}_y\text{:H}_z$ and $\text{Si}_x\text{N}_y\text{:H}_z$ /TCO layers have been measured using UV/VIS spectrometer. The gas flow rate ratio of N_2/SiH_4 has been varied from 235 to 470. Decrease in transmittance of $\text{Si}_x\text{N}_y\text{:H}_z$ /TCO layer is observed with increase in silicon concentration in the film. Refractive index of $\text{Si}_x\text{N}_y\text{:H}_z$ also increased with an increase of the silicon content in the films. The reflectance of TCO films has been decreased from 15% to 8% when $\text{Si}_x\text{N}_y\text{:H}_z$ film is incorporated between glass substrate and TCO film. An improvement of around 20% has been observed in current density of solar cells having $\text{Si}_x\text{N}_y\text{:H}_z$ film as refractive index matching layer with refractive index 1.83. Thus, $\text{Si}_x\text{N}_y\text{:H}_z$ film as refractive index matching layer can be used to improve the solar cells device efficiency. © 2013 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4807609>]

I. INTRODUCTION

Carbon emission free energy technology is the demand of current time due to increasing price of fossil fuel. Crystalline silicon wafer based photovoltaic technology is now stabilized but its production cost seems difficult to reduce. While thin film based silicon solar cells have ability to fabricate on glass as well as on flexible substrates with low module cost. Hydrogenated amorphous silicon is the most promising material due to its high absorption coefficient and low processing temperature, but light-induced degradation of initial efficiency requires further improvement in device structure.¹ Light trapping scheme using textured transparent conducting oxide (TCO) surface is used to reduce reflection of light from front side and

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thus improvement in device is reported.^{2,3} Another light management scheme of better choice is application of index matching layer between glass and TCO layer. The glass has refractive index ~ 1.5 and TCO has ~ 2.0 , so use of a thin layer which has refractive index less than 2.0 and more than 1.5 can be used between them to collect more photons.

Plasma deposited amorphous silicon nitride ($\text{Si}_x\text{N}_y\text{:H}_z$) thin films have been used as passivation layer for crystalline silicon solar cells, thin film transistors, and as a top anti-reflection coating on silicon solar cells.^{5,6} Recently, silicon nitride films have been the object of attention as an index-matching layer between glass substrate and TCO layer as it has a tunable optical band gap from 5 eV to 1.78 eV.^{4,7} Silicon nitride films deposited by plasma enhanced chemical vapour deposition (PECVD) require either silane/ammonia or silane/nitrogen gas mixture as process gas. Much research is done on silane/ammonia based silicon nitride films. However, some work has been reported in the literature on silicon nitride where economically and environmentally safe nitrogen is preferred instead of toxic and hazardous ammonia.⁸ Thus, a green chemistry route paved the way to deposit silicon nitride films using silane/nitrogen gas mixture.

For amorphous silicon solar cells application, our paper demonstrates the effect of nitrogen to silane gas flow rate ratio on growth conditions, antireflection properties, refractive index, and chemical structure of amorphous $\text{Si}_x\text{N}_y\text{:H}_z$ films deposited by PECVD. All films have been characterized by Fourier Transform Infrared (FTIR) spectroscopy, UV-visible spectrophotometer, and spectroscopic ellipsometry. Amorphous silicon solar cells using $\text{Si}_x\text{N}_y\text{:H}_z$ as index matching layer between glass and TCO layer are fabricated and characterized.

II. EXPERIMENTAL DETAILS

Hydrogenated $\text{Si}_x\text{N}_y\text{:H}_z$ films with varying nitrogen to silane ratio have been deposited on glass and silicon substrates using a RF-PECVD system (OXFORD Plasma Lab System-100) operating at a frequency of 13.56 MHz. All the substrates were ultrasonically cleaned using de-ionized water, acetone, and ethyl alcohol respectively. Silicon substrates were further rinsed in diluted HF solution. Process pressure, substrate temperature, and RF power have been kept at 600 mTorr, 300 °C, and 40 W respectively. Silane and nitrogen gas mixture with different SiH_4 flow rate was used to change the stoichiometry and refractive index of $\text{Si}_x\text{N}_y\text{:H}_z$ films. The flow rate of N_2 has been kept at 940 sccm and SiH_4 flow rate was decreased from 4 sccm to 2 sccm. ZnO:Al (AZO) was used as TCO due to its high transparency and resistance to gaseous plasma. Al doped ZnO thin films as TCO layer have been deposited by pulsed DC magnetron sputtering unit. AZO films were deposited at 200 W power, 150 KHz frequency, and 0.05 mbar deposition pressure. Ar gas was used as process gas with 15 sccm flow rate. The substrate temperature was kept at 200 °C during deposition of AZO films. Before solar cell fabrication, 800 nm thin film of AZO was deposited on both bare glass substrates and SiN layer coated glass substrates. AZO will act as anode for solar cells. Amorphous silicon (a-Si:H) solar cells have been fabricated with p-i-n structure followed by Al deposition as back electrode.

Amorphous silicon layers were deposited using RF-PECVD system operating at a frequency of 13.56 MHz. Process pressure, substrate temperature, and RF power were kept at 1000 mTorr, 250 °C, and 10 W respectively. Silane gas in Ar dilution was used as process gas. For device fabrication, first 20 nm p-type a-Si:H thin film was deposited using di-borane (B_2H_6) with SiH_4 and Ar as feed gas. Then, main active layer a-Si:H was deposited using pure silane gas with a flow rate of 25 sccm and Ar gas with a flow rate of 1000 sccm. The thickness of main absorbing layer was 350 nm. The n-type a-Si:H layer was deposited using phosphine gas with SiH_4 and Ar as feed gas. The thickness of n-a-Si:H film was 30 nm. After deposition of p-i-n layers, substrates were taken out from PECVD chamber and transferred to thermal evaporation system for depositing back electrode. Under high vacuum condition ($\sim 6 \times 10^{-6}$ mbar pressure), 200 nm thin Al film was thermally evaporated on p-i-n structure using metal mask. Current-voltage (I-V) measurements for all devices have been performed under a simulated AM1.5 spectrum using Newport Xenon lamp.

UV-visible transmittance and reflectance of all films have been measured using spectrometer (Perkin-Elmer UV/VIS Lambda 750) in the wavelength range of 300–900 nm. Using

transmittance data, the optical band gap of $\text{Si}_x\text{N}_y\text{H}_z$ films was calculated by Tauc method. FTIR spectra were taken in the wave-number range of 400 to 4000 cm^{-1} in transmission mode using Bruker Alpha-T System. Refractive index and thickness of films were measured using spectroscopic ellipsometer (J.A. Woollam). The measured data were analyzed using completeEASE software. A four phase model was used to describe the material system, i.e., air/roughness/SiN/SiO₂/Si substrate, where Si substrate thickness was treated as infinite, SiO₂ thickness was fixed at 2 nm, and thickness of SiN was varied from 400 to 600 nm. The measured data were fitted for different layers of model using Bruggeman effective medium approximation, Tauc-Lorentz model, and Sellmeier expression as described by Jellison and Modine.⁹

III. RESULTS AND DISCUSSION

FTIR spectroscopic technique has been used for chemical analysis of $\text{Si}_x\text{N}_y\text{H}_z$ films. The chemical bonds in $\text{Si}_x\text{N}_y\text{H}_z$ are detected by measuring the radiation absorption bands in the near infrared range. FTIR spectroscopy has been used to determine the nature of Si-N, Si-H, and N-H bonding in the film. The FTIR spectra of $\text{Si}_x\text{N}_y\text{H}_z$ film are shown in Fig. 1. In FTIR spectra of $\text{Si}_x\text{N}_y\text{H}_z$ films, peaks at 650–670, 760–900, 925, 1065, 1260, 1530, and 2052 cm^{-1} were detected. The absorption peak at 650 cm^{-1} is due to $\text{N}_3\text{-SiH}$ bond in $\text{Si}_x\text{N}_y\text{H}_z$ film. Peak at 670 cm^{-1} is due to Si-NH_2 bond. Peak between 760 and 950 cm^{-1} is due to Si-N stretching mode.¹⁰ The shift from standard peak position is because of the effect of neighboring atoms on frequencies of its mode. The intensity of Si-N bond peak is higher for lower SiH_4 gas flow rate, i.e., higher N_2/SiH_4 flow rate ratio. The absorption peak at 925 cm^{-1} is due to deformation vibration of Si-H₂ bond. The absorption peak at 1065 cm^{-1} is due to symmetric stretching mode of $\text{N}_3\text{-Si-H}$ group. The N-H bond peak is observed at 1260 cm^{-1} and N-H₂ deformation peak at 1530 cm^{-1} . The absorption peak at 2100 cm^{-1} is due to H-Si-NSi₂ group. The stretching mode for Si-H bonds is observed at 2052 cm^{-1} .

Broad peak from 2800 cm^{-1} to 3500 cm^{-1} corresponds to peculiarities of stretching mode for N-H modes. The intensity of peak of N-H bond reduces with increasing N_2/SiH_4 flow rate ratio because lower hydrogen atoms are available for making N-H bond.^{11,12}

Fig. 2 shows the reflectance of $\text{Si}_x\text{N}_y\text{H}_z$ films with different N_2/SiH_4 flow rate ratios. It is observed from the figure that as N_2/SiH_4 flow rate ratio decreased from 470 to 235, the percentage reflectance of films also decreased from 3.45% to 0.9% at 500 nm wavelength. Fig. 3 shows transmittance and reflectance of 800 nm thin ZnO:Al films deposited on $\text{Si}_x\text{N}_y\text{H}_z$ layer of three different thicknesses (SN1 = 597 nm, SN2 = 446 nm, and SN3 = 435 nm). The film thickness of $\text{Si}_x\text{N}_y\text{H}_z$ films was decreased with decreasing the N_2/SiH_4 flow rate ratio. The observed

FIG. 1. FTIR spectra of $\text{Si}_x\text{N}_y\text{H}_z$ films with different N_2/SiH_4 flow rate ratios.

FIG. 2. Reflectance of $\text{Si}_x\text{N}_y\text{:H}_z$ films with different N_2/SiH_4 flow rate ratios and ratio is denoted by SN1, SN2, and SN3.

reduction in deposition rate, with decreasing N_2/SiH_4 flow rate, may be due to less availability of “N” radicals near the substrate for reaction. It was observed from the figure that reflection of $\text{Si}_x\text{N}_y\text{:H}_z$ films reduces with decreasing N_2/SiH_4 flow rate ratio. When $\text{Si}_x\text{N}_y\text{:H}_z$ film layer is used between glass and TCO layer as index matching layer, the reflectance of TCO layer decreased from 15% to 8%.

Lowering of reflectance of TCO coated glass means more availability of light for absorber layer of solar cells. The reflectance of $\text{ZnO:Al}/\text{Si}_x\text{N}_y\text{:H}_z/\text{glass}$ combination is 7% lower than $\text{ZnO:Al}/\text{glass}$. Fig. 4 shows the refractive index of $\text{Si}_x\text{N}_y\text{:H}_z$ films with different N_2/SiH_4 flow rate ratios. Refractive index of films decreased with decreasing silane flow rate with respect to constant flow rate of nitrogen, which means more availability of nitrogen to make Si-N bonds as confirmed by FTIR spectra of films. At higher N_2/SiH_4 flow rate ratio, the feed rate of “N” radicals near the substrate will be high and thus more number of “N” radicals will incorporate in the film. Thus, high N-content and low Si-content films become porous, which causes lowering of refractive index. $\text{Si}_x\text{N}_y\text{:H}_z$ films deposited with N_2/SiH_4 flow rate ratio of 470 showed least refractive index of 1.83 at a wavelength of 640 nm. $\text{Si}_x\text{N}_y\text{:H}_z$ layer with N_2/SiH_4 flow rate ratio of 470 is used as index matching layer for p-i-n solar cells.

FIG. 3. Transmittance and reflectance of $\text{Si}_x\text{N}_y\text{:H}_z$ films with different N_2/SiH_4 flow rate ratio denoted by SN1, SN2, and SN3.

FIG. 4. Refractive index of $Si_xN_y:H_z$ films measured at wavelength of 640nm with different N_2/SiH_4 flow rate ratio denoted by SN1, SN2, and SN3.

Both types of silicon solar cells, having SiN as index matching layer and without SiN, were fabricated in order to evaluate the effect of index matching layer between glass and TCO. Amorphous silicon solar cells have been fabricated with device structure {Al(200 nm)/n-a-Si:H(30 nm)/i-a-Si:H(350 nm)/p-a-Si:H(20 nm)/TCO(800 nm)/SiN/glass} as shown in Fig. 5. The I-V characteristics of a-Si:H thin film solar cell with and without $Si_xN_y:H_z$ layer have been measured. Solar cells incorporating SiN as index matching layer show a large gain in current. Current density increased from 5.75 mA/cm² to 7.46 mA/cm² and efficiency increased from 1.89% to 2.08% after application of $Si_xN_y:H_z$ as index matching layer in device structure. The measured I-V characteristics of a-Si:H thin film solar cells are shown in Fig. 6. The improvement in efficiency of solar cells was relatively low because of poor fill factor (FF) in both types of solar cells. Solar cells containing SiN as index matching layer show slight reduction of open circuit voltage (V_{oc}) and FF, but this leads to a minor difference in cell efficiency. V_{oc} of cells decreased from 0.83 V to 0.80 V and FF decreased from 0.39 to 0.35. Open circuit voltage (V_{oc}) generally depends on the difference in energy levels of the electron donor and acceptor layer. The reason behind slight decrease in V_{oc} may be due to dominant role played by charge carriers.

FF of solar cell is defined as the ratio of area of maximum rectangle fitted in I-V curve ($V_m \cdot I_m$) and the product of V_{oc} and I_{sc} . The ideal FF is free from losses due to recombination

FIG. 5. Device structure of amorphous silicon solar cell using $Si_xN_y:H_z$ film as index matching layer.

FIG. 6. I-V characteristics of a-Si:H thin film solar cell with and without $\text{Si}_x\text{N}_y\text{H}_z$ layer.

of charge carriers and series resistance. The possible reasons for poor FF are poor interface between active layer and electrodes, traps inside active layer, and non-uniformity of TCO layer. The FF loss has been calculated using equation ($\text{FF loss} = I_{\text{sc}} \cdot V_{\text{oc}} - I_m \cdot V_m$), where V_m is voltage at the maximum power point, I_m is current at maximum power point, V_{oc} is open circuit voltage, and I_{sc} short circuit current.¹³ FF loss is increased from 0.00295 to 0.0039 in devices which incorporated SiN. This FF loss is comparatively very low in both types of devices but any loss in FF will reduce the efficiency of the solar cells. Another factor which affects the FF of solar cells is dark leakage current.¹⁴ In our devices, even though both types of solar cells are fabricated in identical conditions still variation in V_{oc} and FF is observed.¹⁵ This reduction in FF of solar cells incorporating SiN, may be due to higher leakage current. Another possibility of lower FF is non-uniformity of TCO surface. It is reported that deposition of SiN films using silane/nitrogen gas mixture causes high surface roughness due to a very high surface reactivity of the N atoms.¹⁶ This high surface roughness of SiN films may create non-uniform TCO layer, which also affects the properties of p-layer and thus causes reduction in FF of solar cells.

IV. CONCLUSIONS

A systematic analysis for optimizing the refractive index of $\text{Si}_x\text{N}_y\text{H}_z$ films for index matching layer application is demonstrated. $\text{Si}_x\text{N}_y\text{H}_z$ layer with refractive index of 1.83 has been used as index matching layer for solar cells application. The current density of a-Si:H thin film solar cells increased from 5.75 to 7.46 mA/cm^2 after application of $\text{Si}_x\text{N}_y\text{H}_z$ index matching layer between glass and TCO.

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