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Broad range refractive index engineering of Si_xN_y and SiO_xN_y thin films and exploring their potential applications in crystalline silicon solar cells

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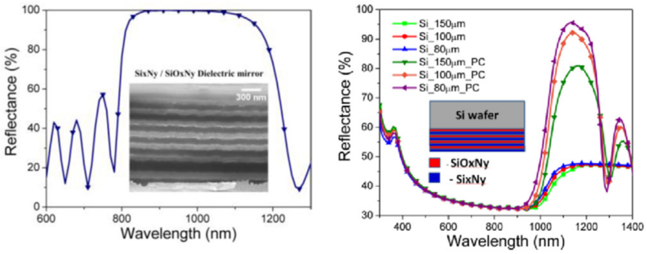
HIGHLIGHTS

- Silicon rich Si_xN_y & SiO_xN_y films are fabricated by low temperature rf-PECVD.
- The refractive indices of these films could be varied from 1.53 to 3.29.
- Different applications of these films in solar cells are demonstrated.
- The films are used to fabricate dielectric Bragg mirror, multilayer & rugate ARC.
- Dielectric mirror acts as the back reflector with excellent surface passivation.

GRAPHICAL ABSTRACT



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ABSTRACT

Silicon nitride (Si_xN_y) and silicon oxynitride (SiO_xN_y) are materials that can find multifarious application in solar cells by tuning their microstructure and optical properties. In this work we have investigated a series of Si_xN_y and SiO_xN_y films deposited by rf-PECVD at a low temperature of 200 °C having different compositional and optical properties by changing the gas mixture ratio during deposition resulting in varying refractive index materials. The effect of varying gas ratio on the composition of these materials has been investigated comprehensively by X-ray Photoelectron Spectroscopy (XPS) and the bond analysis using Fourier Transform Infrared Spectroscopy (FTIR) thereby creating a material library to select materials of desired properties and bond composition for specific applications in solar cells. The structural and optical properties were also analysed using Micro-Raman spectroscopy, X-ray Diffraction (XRD), UV-VIS-NIR spectrophotometer and ellipsometry. The films were found to be amorphous, hydrogenated, compact, presents high deposition rates and needs lesser thermal budget. In order to demonstrate their multifaceted application potential, we have fabricated multilayer anti-reflection coatings (ARC), novel Rugate ARC, dielectric Bragg mirrors using SiO_xN_y/Si_xN_y multi layers as back reflectors for thin crystalline silicon (c-Si) wafers as well as the passivation of c-Si. The novel bilayer passivation stack over CZ p-type c-Si wafers using SiO_xN_y/Si_xN_y presented a minority carrier lifetime of 169 µs with an implied V_{oc} of 673 mV.

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1. Introduction

Stoichiometric silicon nitride (Si_3N_4) films find many

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applications in semiconductor industry and are used extensively in Microelectronics and solar cells. Si_3N_4 films are used as oxidation barriers, dielectric, adhesion layer and encapsulant in Integrated Circuit technology preventing the contamination of IC's from alkaline ions & moisture. The nearly stoichiometric hydrogenated Si_3N_4 by plasma deposition with a refractive index ≈2 is the state-of-art antireflection coating for the p-type silicon wafer solar cells. Moreover Si_3N_4 provides excellent surface passivation for n-type silicon and high amount of hydrogen present in this films facilitate the bulk passivation. Silicon rich silicon nitride thin films would have higher refractive indices and are not explored much on the microstructural properties and for their application possibility. Though silicon oxynitride finds application as insulation interlayer and surface passivation in microelectronics, it is a relatively new material in photovoltaics. Silicon oxynitride can be used as the passivation layer for both n and p-type silicon [1], possess high thermal stability [2] and high transparency which makes them attractive to be used in multilayer antireflection coatings with high photon penetration and as highly transparent rear surface passivation layer in wafer silicon solar cells. There has been an increased research interest in PV for silicon oxynitride due to its potential to be a good passivation layer since they are hydrogenated, more compact as compared to PECVD SiO_2 and are industrially viable due to its high deposition rates. In this paper we highlight the fabrication and characterization of a series of tunable silicon rich silicon nitrides (Si_xN_y) and silicon oxynitrides (SiO_xN_y) thin films deviated from Stoichiometric Si_3N_4 by changing the gas ratio such that their properties can be tailored based on different applications in photovoltaics. In the article we have also demonstrated multilayer ARC using these varying refractive indices Si_xN_y and SiO_xN_y thin films for crystalline silicon solar cells. We have introduced a novel ARC structure for crystalline Si solar cells called as Rugate ARC using Si_xN_y and SiO_xN_y which helps to have reduced thickness and additionally stated the experimental challenges involved for varying refractive index layers. In our previous work we had developed one dimensional photonic crystal [3] [4] using Si_xN_y and SiO_xN_y thin films. We take it one step further to trap infrared photons for thin crystalline silicon wafers by acting as a bragg reflector at the rear end. Si_xN_y having refractive index till 2.4 has been shown to act as good passivation layer for front n type emitter. However the fixed oxide charge has to be controlled for rear side [5] which limits its application as back surface passivation layer. SiO_xN_y could be a better passivation layer for rear surface due to the reasons mentioned previously and we have explored the passivation quality of our SiO_xN_y films. This will help us to develop a photonic crystal back reflector in future which will serve the dual purpose of light trapping to increase the short circuit current and rear passivation to improve the open circuit voltage of the solar cell.

We furthermore present the characterisation of varying refractive index silicon rich silicon nitride and silicon oxynitride films deposited by rf-PECVD by varying the gas flow ratios during the deposition at a low temperature of 200 °C, thereby decreasing the thermal budget of the process. The low temperature process of these thin films further increases their application in flexible devices over polymer substrates. Unlike sputtering, deposition by PECVD decreases the plasma damage caused to the layers underneath and also provides higher deposition rates. It also helps to precisely control the properties of the materials by controlling the growth parameters thus aiding reproducibility of the layers. PECVD is already an existing industrial process for commercial solar cells and hence these materials can find direct application in the inline cell production without any additional equipment cost.

Since these materials find extensive use in the photovoltaic industry we have developed a material library of nitrides and oxynitrides deviated from their stoichiometry to have different compositional and optical properties so that the material could be selected based on the requirement of the application. This detailed study could be useful for depositing a series of silicon oxynitrides for various applications in microelectronics as well. A systematic analysis of the optical, structural, composition and bond analysis of these materials has been provided in this paper in order to facilitate their application in novel structures for photovoltaics. A detailed bond analysis of all the films has been carried out since an understanding of the bond configuration provides important information with regard to the stability, passivation quality, etc. In order to demonstrate the manifold application of these films in photovoltaics, we have used different refractive index materials to design multilayer ARCs for c-Si solar cells, novel passivation of p-type silicon using silicon oxynitride/silicon nitride stack and also designed and fabricated a distributed Bragg dielectric back reflector over 80,100 and 150 μm thick c-Si wafers.

2. Experimental details

Si_xN_y and SiO_xN_y thin films are deposited by radio frequency parallel plate PECVD system, Plasmalab 100 by Oxford Instruments. The films are deposited at a pressure of 700 mtorr, substrate temperature of 200 °C and a plasma power density of 60 mW/cm^2. The rf power is applied at the upper electrode and the samples are placed over the lower electrode which is grounded. The substrate temperature is given by a heater below the grounded electrode. Si_xN_y is deposited using Silane (SiH_4) and Ammonia (NH_3) as the precursor gas. NH_3 is preferred over N_2 for Si_xN_y due to lower dissociation energy of NH_3 as compared to N_2 since the dissociation energy of N—H bond is lesser than that of N—N bond as explained by Lee et al. [6]. In order to have films of varying refractive index, the silicon nitride is made silicon rich by decreasing the NH_3 to SiH_4 ratio from 1 to 0.0625. SiO_xN_y films are deposited using SiH_4, Hydrogen (H_2) and Nitrous Oxide (N_2O) as the precursors. SiH_4 to H_2 gas flow ratio is maintained constant at 0.75, whereas the SiH_4 to N_2O ratio is varied from 0.5 to 0.11 to increase the oxygen content in SiO_xN_y and thereby decreasing the refractive index [7]. The deposition rate of Si_xN_y varies between 0.4 nm/s to 0.92 nm/s and that of SiO_xN_y varies between 0.22 nm/s to 1.1 nm/s for different gas mixture ratios. In Si_xN_y the deposition rate increases when NH_3 to SiH_4 ratio decreases, whereas for SiO_xN_y the deposition rate increases when N_2O flow rate increases. In order to uniquely identify the materials, we have used gas mixture ratios R_{SiN} and R_{SiON}, for Si_xN_y and SiO_xN_y films respectively, which are defined as:

$$R_{SiN} = \frac{\varnothing_{NH_3}}{\varnothing_{SiH_4}} \tag{1}$$

and

$$R_{SiON} = \frac{\varnothing_{N_2O}}{\varnothing_{N_2O} + \varnothing_{SiH_4}} \tag{2}$$

where, $\varnothing_{NH_3}$, $\varnothing_{SiH_4}$ and $\varnothing_{N_2O}$ are the gas flows of NH_3, SiH_4 and N_2O respectively in sccm. The gas mixture ratios for the deposition of Si_xN_y and SiO_xN_y films are given in Tables 1 and 2 respectively. The sample names of the silicon nitride and silicon oxynitride films discussed in this paper are represented as $Si_xN_y\_R_{SiN}$ and $SiO_xN_y\_R_{SiON}$.

The optical, structural and compositional characterizations of the films are performed using different analytical techniques. The Fourier Transform Infrared Spectroscopy (FTIR), reflectometry and X-ray Photoelectron Spectroscopy (XPS) characterization were done with the films deposited on p type CZ Si wafers having orientation <100>, resistivity of 4–7 Ω-cm and thickness of

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