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Development of amorphous carbon-based variable optical gap semiconductor materials

Kensuke HONDA, Kohsuke YOSHINAGA, Akira NAKAHARA

Graduate school of Science and Engineering, Yamaguchi University
1677-1 Yoshida, Yamaguchi-Shi, Yamaguchi, 753-8512, Japan

1. Introduction

The widespread use of a low-cost (7 yen/kWh) and high efficiency (40%) solar cell can contribute to reducing CO_2 emission from fossil fuel use, which leads to global warming. Tandem-type solar cell composed of stacked layers of variable optical gap carbon-based semiconductors can achieve high efficiency solar cell at lower cost. High performance semiconductor with variable optical gap, however, has not been realized by using carbon based materials in the previous studies. The objective of this study is to develop p- and n-type carbon-based semiconductors with variable optical gap ranging from 1.0 eV to 2.5 eV and high semiconducting properties applicable to electronic devices.

In previous publication, the optical gaps of amorphous carbon (a-C) depend on sp^2/sp^3 ratios and the size of sp^2 clusters in a-C. By establishing the method to control sp^2/sp^3 ratios and the size of sp^2 clusters, a-C based variable optical gap semiconductor can be realized. Recently, our research group has successfully synthesized high optical gap (1.76 eV) amorphous carbon semiconductor using plasma-enhanced chemical vapor deposition (PeCVD) method by introducing Si atoms in a-C (Si:C = 1:4) and reducing the size of sp^2 clusters. The ratio of sp^3/sp^2 hybrids can be easily controlled by changing Si/C ratio in a-C films. It results in varying the optical gap of a-C films.

In our method, a liquid source composed of hydrocarbons including impurity (nitrogen) atoms and Si was employed as source material of PeCVD. The atomic ratios of Si and C atoms in Si-added a-C can be easily controlled by changing the composition (Si:C) of the liquid source. In this study, a-C semiconductor with wider optical gap (ca. 2.5 eV) and higher mobility (volume resistivity is ca. 10^1 Ω cm) was tried to be synthesized to realize a-C based variable optical gap semiconductors.

2. Experimental

Si-added nitrogen-doped amorphous silicon carbide films (Si+N-doped a-C) were fabricated by anode-coupling type radio frequency plasma enhanced chemical vapor deposition system. (RF-PeCVD) (13.56 MHz, SAMCO Co., Ltd. Model BPD-1) N-doped a-SiC films were deposited on highly Sb-doped silicon (111) substrate (0.02 Ω cm of resistivity) and insulating glass plate. As a liquid source, mixed solution of tetramethylsilane (TES) (Si and C sources) and 1,1,1,3,3,3-hexamethyldisilazane (HMDS) (Si, C and N sources) at a ratio of [HMDS]/[HMDS]+[TES] = 0.01 was used. Vaporized liquid source was introduced into a reaction chamber by heating at 80°C. The source gas at a flow rate of 1 sccm and H_2 gas at a rate of 150 sccm were introduced simultaneously. Si+N-doped a-C thin films were deposited using plasma at RF power of 150 W. Deposition time was 40 minutes. Typical thickness of the deposited thin films was 2.70 µm. Chemical compositions and optical and semiconductor properties of the resulting Si+N-doped a-C films were examined.

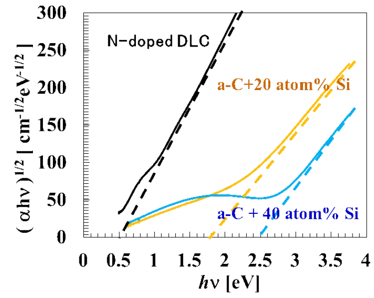

Figure 1. Tauc plot of Si+N-doped a-C

3. Results and discussion

3.1 Chemical composition

The surface of Si+N-doped a-C film deposited on glass was blue in color and highly transparent. It is clarified that the film was composed of silicon and carbon (Si:C:N = 41:56:3) from the results of XPS measurement. The peaks attributable to Si bonded to C and attributable to Si bonded to N were observed in Si 2p peak in XPS spectra, which indicated that Si atoms were dispersed widely and formed the bonding to carbon by sp^3-hybrid orbitals. The atom% of N (donor) in a-SiC was 3 atom%.

3.2 Optical property

Optical gap of Si+N-doped a-C was estimated to be 2.76 eV from Tauc plot of UV-Visible absorption spectra. This value was 1.10 times higher than the target (2.5 eV) (Figure 1). This higher value is thought to be caused by shorten carbon content in the film.

3.3 Semiconductor property

From the results of Hall measurement of Si+N-doped a-C, the type of conduction was n-type, carrier density was 1.78×10^{14} cm^{-3}, and volume resistivity was 1.26×10^3 Ω cm.

The performance of photo-electron conversion at Si+N-doped a-C was examined by photo-electrochemical method (Figure 2) to verify the applicability to photo voltaic system. The value of current with exposure to Hg-Xe lamp (photocurrent) at Si+N-doped a-C was achieved to be 20.6 µA cm^{-2} at 9 V. (Figure 2) This value was 1/8 lower than that obtained at anatase-type titanium dioxide thin films prepared by sol-gel method. Quantum yield of the film was calculated using the value of photocurrent. The quantum yield of photo-excited electron at Si+N-doped a-C was calculated to be 1.46 % using 365 nm of wavelength and 11.8 W cm^{-2} of intensity of Hg-Xe lamp (photons were completely absorbed in the film).

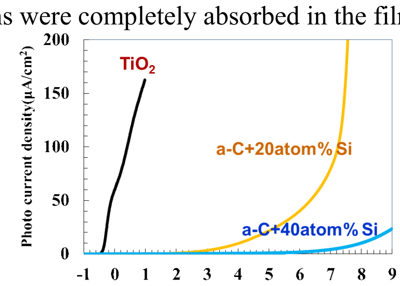

Figure 2. Current-voltage curves with exposure to Hg-Xe lamp of N-doped a-SiC

4. Conclusion

The high performance n-type a-C semiconductor films with variable optical gaps ranging from 1.76 eV to 2.76 eV, which can be utilized to Tandem-type solar cell, could be successfully fabricated. However, it is necessary to increase conductivity in order to apply the film to a solar cell. Higher conductivity will be attempted by incorporating other impurity atoms like P that does not form insulating components such as SiN.