Development of amorphous carbon-based variable optical gap semiconductor materials

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1. Introduction

The widespread use of a low-cost (7 yen/kWh) and high efficiency (40%) solar cell can contribute to reducing CO2 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-applicable to electronic devices.

In previous publication, the optical gaps of amorphous carbon (a-C) depend on sp3/sp2 ratios and the size of sp3 clusters in a-C. By establishing the method to control sp3/sp2 ratios and the size of sp3 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 sp3 clusters. The ratio of sp3/sp2 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) than a-Si:H can be synthesized to realize a-C based variable optical gap semiconductor.

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 Si-doped silicon (111) substrate (0.02 Ω cm of resistivity) and insulating glass plate. As a liquid source, mixed solution of tetramethylsilane (TMS) (Si and C sources) and 1,1,1,3,3-hexamethyldisilazane (HMDS) (Si, C and N sources) at a ratio of [HMDS]/[HMDS]+[TMS] = 0.01 was used. Vaporized liquid source was introduced into a flow reactor, heated at 80°C, into a reaction chamber by heating at 80°C. 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 H2 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.

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 sp3-hybrid orbitals. The atom% of N (donor) in a-SiC was 3 atom%.

3.2 Optical 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 × 1014 cm-3, and volume resistivity was 1.26 × 105 Ω cm.

The quantum yield of photo-excited electron at Si+N-doped a-C was examined by photo-electrochemical method (Figure 2) to verify the applicability to photovoltaic system. The value of quantum yield was 1.10 times higher than the target (2.5 eV) (Figure 1). This higher value is thought to be caused by shortened carbon content in the film.

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.