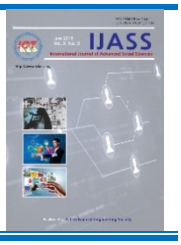
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Solid Carbon Source based Fe-doped Graphene/Amorphous Carbon Hybrid Films for Solar Cell

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Abstract

The Fe-doped graphene/amorphous hybrid carbon films have been synthesized by microwave surface wave plasma chemical vapor deposition (MW-SWP-CVD) on n-type silicon substrate for the application of the photovoltaic cell. Ferric acetylacetonate ($C_{15}H_{21}FeO_6$) is used as a solid carbon source as well as dopant. The characterization of deposited films were studied by Raman spectroscopy, X-ray photoelectron spectroscopy (XPS), and Solar simulator to investigate chemical, structural and electrical properties of the a-C films. Raman spectroscopy shows that the grown film consists of both graphene and a-C. The iron (Fe) contain in the film is confirmed by XPS. The formation of hetero-junction of fabricated device is confirmed by measuring the current density-voltage characteristic, which shows rectifying characteristic under no illumination. The energy conversion efficiency of the device is 2.06 % under the illumination (A.M 1.5, 100 mW/cm^2, 25 ℃).

Keywords: *Amorphous, Carbon, Graphene, MW-SWP-CVD, Silicon, Solar Cell.*

1. Introduction

Carbon is a remarkable element in the earth's crust, existing in many allotropes. The different combinations in which carbon atoms can be arranged produce a wide variety of compounds that have unique and interesting physical, chemical, and electronic properties. Depending on the chemical nature there are varieties of stable forms of carbon ranging from insulator diamond to metallic graphite to conducting/semi-conducting nanotubes. Among various application of carbon materials (such as amorphous carbon (a-C), fullerenes, graphene, and carbon nanotubes), recent study of solar cells are quite interesting [1]. Their interesting optical properties and low cost make a-C films as potential material for solar cell. The a-C films were earliest carbon materials partially replacing silicon (Si) in Si-based solar cells, but their poor conductivity hinders their practical application [2-3]. Excellent conductivity, good transparency, and high mobility make graphene as promising candidate in photovoltaic devices [4]. Lots of studies has been carried out on fabrication of graphene/n-Si Schottky junction photovoltaic devices, but the fabrication process of the device is relatively complex because graphene easily cracks [5].The above mention problem can be overcome by a graphene/a-C hybrid structure.

In this paper, iron (Fe) doped graphene/amorphous hybrid carbon (G/a-C:Fe) films grown on n-type silicon by microwave surface wave plasma chemical vapor deposition (MW-SWP-CVD) and its application on solar Cell is

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studied.

2. Experiment Details

The G/a-C:Fe films were deposited on n-type silicon (2- 20 Ohm-cm) substrate by using the MW-SWP-CVD technique. A schematic diagram of MW-SWP-CVD is shown in Fig. 1.

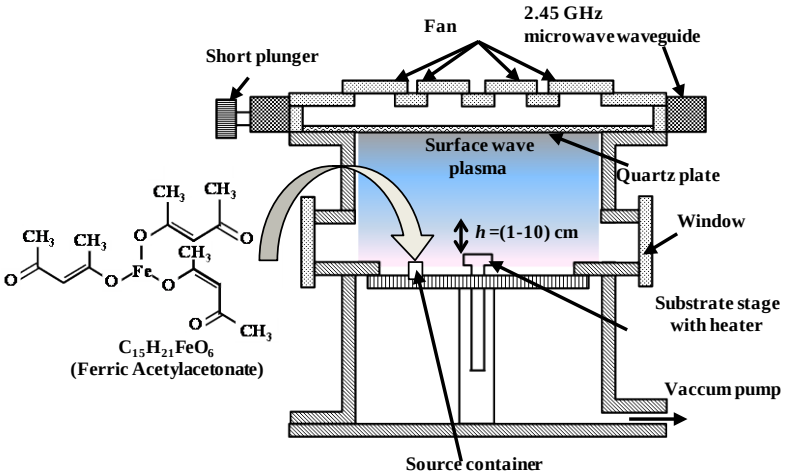


Fig.1. Schematic diagram of SW-MWP-CVD and chemical structure of ferric acetylacetonate.

The surface wave plasma was produce in 40 cm diameter, cylindrical vacuum chamber by introducing a 2.45 GHz microwave through a hollow quartz plate via slot antenna. The maximum microwave power of the system is 2.5 kW. During the surface plasma discharge operation, microwaves damped exponentially below the quartz plate where the electron density (more than 3.6×10^{11} cm^{-3}) generally exceeded the cut-off density of 7.4×10^{11} cm^{-3}. It has been reported that using a hollow quartz plate, plasma can be started smoothly without any density jump and plasma is two times higher than that for a flat plate. High density surface wave plasma can be generated in the meter scale and large area deposition can be achieved at a very fast rate [6].

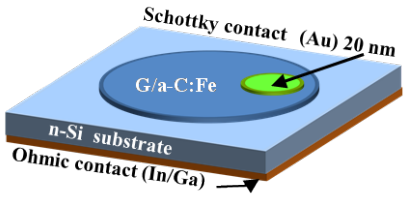


Fig.2. Schematic diagram of photovoltaic device.

The substrates were ultrasonically cleaned in acetone, then methanol, etched in diluted hydrofluoric (HF) solution (10 %) in order to remove the native oxide layer from surface, rinsed in deionized water and dry by N_2 blow. For film deposition, Argon (Ar: 200 sccm) was used as carrier gas and acetylacetonate iron complex ($C_{15}H_{21}FeO_6$) was used as both carbon source as well as dopant. The carbon source is placed in the source container inside CVD chamber as shown in Fig. 1. Before deposition, the chamber was evacuated at 5.0×10^{-4} Pa, and total gas pressure was held fixed at minimum (about 40 Pa) during film deposition. The films were deposited at microwave power of 1200 W for 5 min at stage height of 6 cm and stage temperature of 550 °C. The structure of solar cell device is shown in Fig. 2, which consists of G/a-C:Fe film on n-Si substrate sandwiched between the

Ohmic contact (InGa) and Schottky contact (Au).

The photovoltaic characteristics of the fabricated device were measured with solar simulator. The micro structure and the film compositions of the samples were determined by Raman spectroscopy having laser excitation energy of 532 nm in NRS 300 laser Raman spectrometer and X-ray photoelectron spectroscopy (XPS) in SSX-100.

3. Results and Discussion

3.1. Deposition mechanism

The G/a-C:Fe films were deposited on n-Si substrate by MW-SWP-CVD technique using $C_{15}H_{21}FeO_6$ as solid car-bon source as well as dopant in argon environment. The acetylacetonate iron complex called ferric acetylacetonate can also be written as $Fe(C_5H_7O_2)_3$. The chemical structure of $C_{15}H_{21}FeO_6$ is shown in the left hand side of Fig. 1. The structure consists of discrete molecules linked together in layers by Vander Waals forces. Three acetylacetonate radicals, $(C_5H_70_2)_{-1}$, surround the central iron atom of each molecule, with the oxygen atoms in octahedral coordination [7-8].The boiling point of $C_{15}H_{21}FeO_6$ is about 100 °C. The boiling point of the carbon source and the dopant is very critical to obtain iron doped graphene/amorphous hybrid carbon film using separate source and dopant. Here we have use iron complex as a carbon source and dopant, which overcomes the above problem. As the temperature of chamber increases and crosses the boiling point of $C_{15}H_{21}FeO_6$, evaporation occurs. Then, the evaporated $C_{15}H_{21}FeO_6$ are decomposed into different carbon species and radicals by plasma process. Solid source has also good catalytic decomposition properties at low temperature. Hydrogen and oxygen escape from chamber in the gas form, while Fe and C sticks to Si surface. Hence, G/a-C:Fe films is deposited on Si substrate.

3.2. Raman spectroscopy

Raman spectroscopy provides a quick and facile structural and quality characterization of the material. The Raman studies of G/a-C:Fe film grown on n-Si substrate is presented in Fig. 3. Four peaks are observed at around 1345, 1575, 2666 and 2922 cm^{-1} corresponds to D (due to breathing modes of sp2 carbon atoms in rings), G (due to bond stretching between pairs of sp2 carbon atoms), 2D (second-order zone-boundary photons) and G+D, respectively. The prominent D peak indicates the presence of localized sp3 defects within the sp2 carbon network [9] as well as may due to doping effect and the much stronger interaction between carbon and silicon. The G peak is due to the doubly degenerated zone center E2g mode [10].The intensity of 2D band is dependent on the number of layers [11]. Raman study indicates that the grown film consists of both graphene and a-C (i.e., graphene/amorphous hybrid carbon).

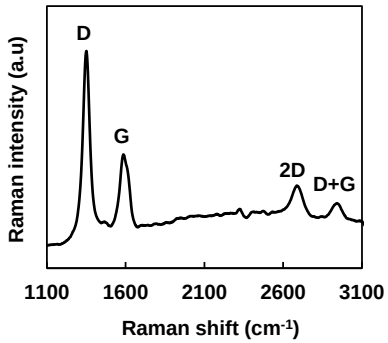


Fig.3. Raman spectroscopy of G/a-C:Fe film grown on n-Si substrate.

3.3. XPS spectroscopy

XPS spectroscopy In order to investigate the content of iron atoms at different depths, core-level XPS were also performed on G/a-C:Fe film grown on n-Si substrate for different etching time (0, 300, 600 sec). Fig. 4(a) shows the wide XPS spectra of G/a-C:Fe film deposited on silicon substrate. It clearly shows the presence of O1s, C1s, and Fe3p peaks at around 532.78, 284.39, and 51.06 eV, respectively. Fig. 4(b) shows the high resolution XPS spectra of G/a-C:Fe films with photoelectron emission from the Fe3p core level peak centered at 51.06 eV corresponding to iron atoms in films. Fig. 4(c) shows the high resolution XPS spectra of G/a-C:Fe films with photoelectron emission from the C1s core level peak centered at 284.39 eV corresponding to sp3/sp2 hybrid carbon. Similarly, high resolution XPS spectra shown in Fig. 4(d) shows the presence O1s peak centered at 284.39 eV corresponds to oxygen in film, which may be due to surface chemi-absorption on the film surface. Others peaks corresponding to Fe2s, Fe3s and Fe2p were also observed in core level XPS spectra.

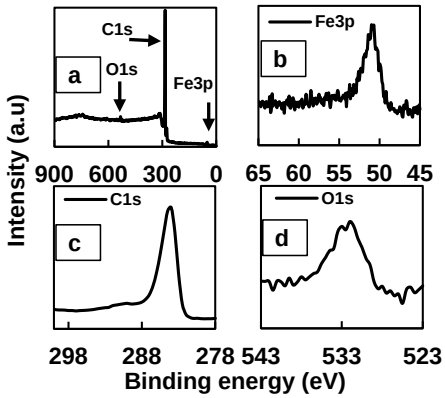


Fig. 4. XPS spectra of G/a-C:Fe film grown on n-Si substrate. (a) Core level XPS spectra indicating carbon, iron, and oxygen, (b) high resolution Fe3p XPS, (c) high resolution C1s XPS, and (d) high resolution O1s XPS.

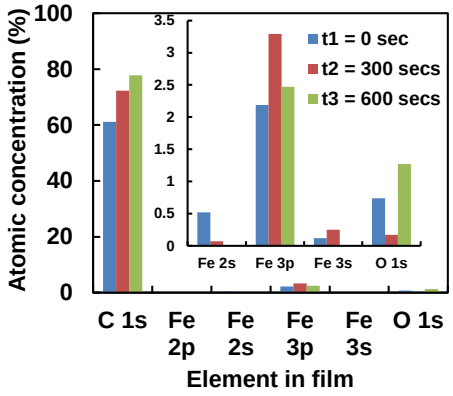


Fig. 5. Atomic concentration versus elements in films measured by XPS.

The quantitative analysis of atomic concentration of different elements (iron, carbon, oxygen) in the films at different depths, which were performed XPS spectroscopy by etching at different time intervals (0, 300, and 600 secs) is shown in Fig. 5. Atomic concentration of all elements except oxygen is higher at nearly middle portion of film compare to surface of film and near substrate. Atomic concentration of Fe3p is higher than that of Fe2p, Fe2s, and Fe3s. The film consists of about nearly 5 % iron atoms in 78 % carbon and remaining are others.

3.4. Photovoltaic characteristic

The current density-voltage characteristic of the fabricated device was measured with solar simulator under illumination (A.M 1.5, 100 mW/cm^2, 25 °C) and dark conditions. The current density-voltage characteristic is shown in Fig. 6. Without illumination, the device shows rectifying characteristic, indicating the formation of hetero-junction. Upon illumination the device shows PV performance with open-circuit voltage (V_{oc}) of 294 mV and short-circuits current density (J_{sc}) of 31 mA/cm^2). The fill factor (FF) and energy conversion efficiency (η) is 22 % and 2.06 %, respectively.

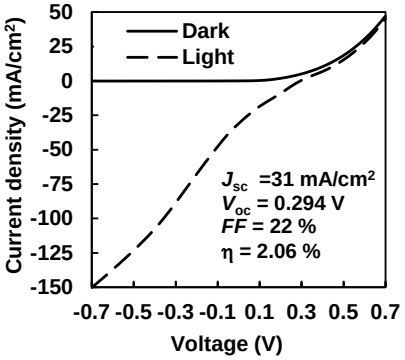


Fig. 6. Current density-voltage characteristic of the fabricated device.

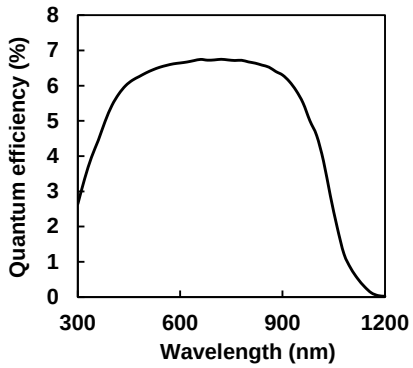


Fig. 7. Quantum Efficiency as a function of wavelength of the fabricated device.

The quantum efficiency (QE) as a function wavelength for the fabricated device is shown in Fig. 7. It shows QE is almost constant in the wide range of wavelength from 300 nm to 1000 nm.

4. Conclusion

The Fe-doped graphene/amorphous hybrid carbon films on n-type silicon substrates were deposited by MW-SWP-CVD using $C_{15}H_{21}FeO_6$ as a solid carbon source as well as dopant. This result indicate the possibility of fabricating solar cell using iron contained solid carbon source, which is confirmed by XPS. It is obtained that the energy conversion efficiency of the device is 2.06 % under the illumination (A.M 1.5, 100 mW/cm^2, 25 °C).

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