



Wide-bandgap nanocrystalline silicon-carbon alloys for photovoltaic applications

Jun-Sik Cho^{a,*}, Eunseok Jang^a, Dongmin Lim^{a,b}, Seungkyu Ahn^a, Jinsu Yoo^a, Ara Cho^a,
Joo Hyung Park^a, Kihwan Kim^a, Bo-Hun Choi^{b,*}

^a Photovoltaic Laboratory, Korea Institute of Energy Research, 152 Gajeong-ro, Yuseong-gu, Daejeon 34129, Republic of Korea

^b Department of Materials Physics, Dong-A University, 840 Saha-gu, Busan 604-714, Republic of Korea

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ABSTRACT

p-Type hydrogenated silicon carbide (p-SiC:H) films were prepared using plasma-enhanced chemical vapor deposition by systematically incorporating carbon into nanocrystalline silicon networks. The carbon content of the films was varied by adjusting the methane/silane (CH_4/SiH_4 ; R_C) ratio. The chemical bonding, microstructural, and electro-optical properties of the films were investigated in detail. With increasing R_C , the silicon-carbon bond density in the films increased gradually and the overall microstructure became amorphous. Through sophisticated control of the carbon content of the films during high hydrogen dilution deposition, p-type hydrogenated nanocrystalline SiC:H (p-nc-SiC:H) films with nanometer-scale Si crystallites embedded in an amorphous SiC:H matrix were obtained. The p-nc-SiC:H films had wide bandgaps larger than 2 eV and reduced parasitic light absorption at wavelengths below 550 nm. When using wide-bandgap p-nc-SiC:H films with superior electro-optical properties as window layers in n-i-p flexible and p-i-n semi-transparent nc-Si:H solar cells, in place of conventional p-nc-Si:H layers with narrow bandgaps, enhanced cell performance was achieved because of both high open circuit voltage (V_{OC}) and high quantum efficiency values at short wavelengths of 350–550 nm. The conversion efficiencies of the flexible nc-Si:H solar cells increased from 6.37% ($V_{OC} = 0.41$ V, $J_{SC} = 22.81$ mA/cm², and FF = 68.10%, where J_{SC} is the short-circuit current density and FF is the fill factor, respectively) to 7.89% ($V_{OC} = 0.51$ V, $J_{SC} = 24.04$ mA/cm², and FF = 64.37%). A further increase in the conversion efficiency to 9.18% was obtained by inserting a very thin, highly-doped p-nc-SiC:H buffer layer between a low-doped p-nc-SiC:H window layer and an indium tin oxide front contact. A significant increase in the conversion efficiency from 3.66% ($V_{OC} = 0.42$ V, $J_{SC} = 14.83$ mA/cm², and FF = 58.69%) to 4.33% ($V_{OC} = 0.45$ V, $J_{SC} = 17.74$ mA/cm², and FF = 54.18%) was achieved with the semi-transparent nc-Si:H solar cell, with an average optical transmittance of 17.29% in the visible wavelength region of 500–800 nm.

1. Introduction

Hydrogenated thin-film silicon (Si) alloys prepared by plasma-enhanced chemical vapor deposition (PECVD) have received attention for use in various optoelectronic applications such as solar cells, light-emitting diodes, color sensors, and phototransistors, because their microstructural, chemical, and electro-optical properties can be widely tuned by controlling deposition parameters [1–5]. In particular, wide-bandgap Si alloys combined with suitable electrical conductivity as p-type or n-type-doped materials are considered key materials for applications in Si-based photovoltaic devices, as window layers for thin-film Si solar cells and as emitter layers for Si heterojunction solar cells [6,7]. These wide-bandgap layers are beneficial for reducing the optical absorption loss of light incident on the solar cells and for increasing the

open circuit voltage (V_{OC}), leading to enhanced cell performance. Among various Si alloys including carbide, oxide, and nitride, hydrogenated amorphous silicon carbide (a-SiC:H) has been widely studied and successfully employed in solar cells, because its wide bandgap can be controlled by changing the ratio of carbon to silicon (C/Si) in the films [8]. The optical bandgap of a-SiC:H films can be modified from about 1.8 eV for a-Si:H and to over 3.5 eV for a-C:H by changing the C/Si ratio. The bandgap of the films increases with increasing carbon content in the structural network of the films; however, the electrical conductivity of a-SiC:H films worsens and structural disorder increases. These effects prohibit charge transport in the films and ultimately limit the device performance. Recently, hydrogenated nanocrystalline silicon-carbide (nc-SiC:H) films deposited under high hydrogen (H_2) dilution have emerged as replacements for conventional a-Si:H and a-

* Corresponding authors.

E-mail addresses: jscho@kier.re.kr (J.-S. Cho), adamchoi@dau.ac.kr (B.-H. Choi).

SiC:H films because they simultaneously provide wide optical bandgaps and improved electrical properties [9–11]. Films of nc-SiC:H are mixed-phase materials consisting of a-SiC:H matrixes that provide a wide bandgap; further nc-Si:H nanocrystallites improve electrical conductivity [12,13]. Although wide-bandgap nc-SiC:H films have been successfully deposited and used in solar cells, an improved understanding of the influence of deposition conditions during PECVD on the structural and electro-optical properties of nc-SiC:H films is necessary. Their impact on the performance of solar cells also needs to be assessed.

In this study, we fabricated boron-doped nc-SiC:H (p-nc-SiC:H) films with wide bandgaps and high electrical conductivities using PECVD. We investigated the dependence of the microstructural, chemical bonding, and electro-optical properties of the films on deposition conditions, focusing on the gas ratio (R_C) of methane (CH_4) to silane (SiH_4). Furthermore, p-nc-SiC:H films deposited with different R_C ratios were used in flexible nanocrystalline Si (nc-Si:H) thin-film and semi-transparent solar cells employed as window layers. The relationship between the material properties of p-nc-SiC:H window layers and the performance of the solar cells was investigated systematically.

2. Experimental details

p-Type SiC:H (p-SiC:H) films were deposited in a PECVD system using an excitation frequency of 13.56 MHz at a substrate temperature of 150 °C with gaseous mixtures of SiH_4 and CH_4 diluted to 50% in H_2 . Diborane (B_2H_6) diluted to 1% in H_2 was used as a dopant gas. The p-layers were deposited on Corning®Eagle 2000™ glasses, p-type (100) single wafers, and 100- μm -thick stainless steel (SUS) substrates for characterizing the chemical, structural, and electro-optical properties. The deposition pressure and RF power density were maintained at 66.7 Pa and 0.2 W/cm², respectively. Typical deposition conditions used for the p-SiC:H films are presented in Table 1.

X-ray diffraction (XRD), Raman spectroscopy, transmission electron microscopy (TEM), and selected area electron diffraction (SAED) were performed to investigate the microstructural characteristics of the p-SiC:H films. The crystalline volume fractions (X_C) of the films, defined as $X_C = (I_{510} + I_{520}) / (I_{480} + I_{510} + I_{520})$, were determined quantitatively by analyzing the Raman signals attributed to amorphous (480 cm⁻¹) and crystalline (510 and 520 cm⁻¹) regions. The chemical bond structures of the deposited films were measured by Fourier transform infrared (FT-IR) spectroscopy. The optical bandgap E_{04} was determined at the absorption coefficient of 10⁴ cm⁻¹. The planar electrical conductivity of the films was obtained from dark conductivity measurements with coplanar aluminum electrodes at room temperature. Vertical charge transport behavior of the samples was investigated by conductive atomic force microscopy (C-AFM).

Flexible nc-Si:H thin-film solar cells with n-i-p configurations and semi-transparent nc-Si:H thin-film solar cells with p-i-n configurations were prepared on SUS substrates with nanotextured ZnO:Al/Ag back-contacts and glass substrates coated with fluorine-doped tin oxide (FTO) films. Intrinsic absorber layers were deposited using a 60-MHz very-high-frequency glow discharge with a 17.1H₂/SiH₄ (R_H) ratio, a deposition pressure of 40.0 Pa, and a substrate temperature of 150 °C. The 30-nm-thick n-type nc-Si:H (n-nc-Si:H) layers were deposited using

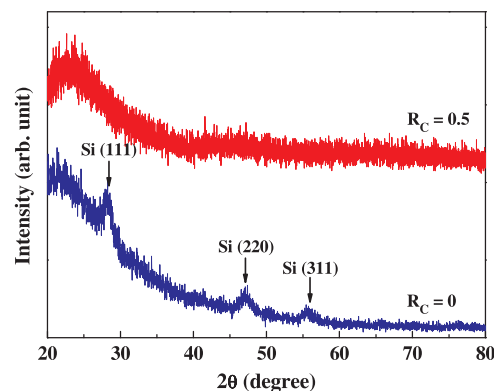


Fig. 1. XRD patterns of p-layers deposited at $R_C = 0$ and 0.5.

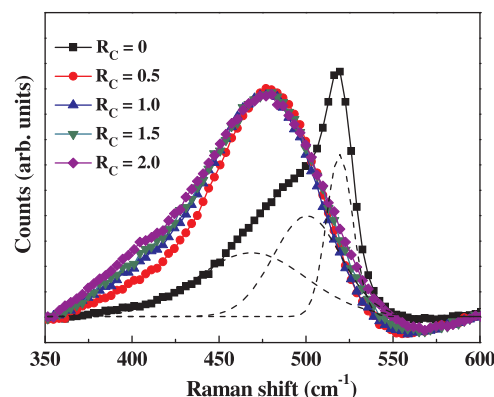


Fig. 2. Raman spectra of p-nc-Si:H and p-SiC:H films deposited as a function of R_C .

a 13.56-MHz radio frequency discharge, and phosphine (PH_3) diluted to 1% H_2 was used as a dopant gas. The area of the solar cells was 2.5 × 2.5 mm², as defined by the geometry of the RF-sputtered indium tin oxide (ITO) front contact. Evaporated Al grids were deposited on the ITO electrodes at room temperature. The structure of solar cells fabricated in this study was SUS substrate/Cr (20 nm)/Ag (300 nm)/ZnO:Al (100 nm)/n-nc-Si:H (30 nm)/i-nc-Si:H (1.0 μm)/p-layer (15 nm)/ITO (80 nm)/Al grid for the n-i-p flexible cell and glass substrate/FTO (600 nm)/p-layer (15 nm)/ i-nc-Si:H (1.0 μm)/ n-nc-Si:H (30 nm)/ ITO (80 nm)/Al grid for the p-i-n semi-transparent cell. Current-voltage (J-V) measurements of the nc-Si:H solar cells with different p-type window layers were performed under an air mass 1.5 illumination with a 100 mW/cm² light intensity at 25 °C using a WACOM WXS-155S-L2. The series resistance (R_s) values of the solar cells were extracted from the J-V curves near open circuit conditions [14]. The spectral response of the samples in the wavelength range of 350–1100 nm was determined from measurements of the external quantum efficiency (EQE). The optical transmittance, absorbance, and reflectance of the semi-transparent solar cells were measured by a UV-Vis-NIR spectrometer equipped with an integrating sphere at wavelengths from 350 to 1100 nm. The average optical transmittance of the semi-transparent solar cells was evaluated from the data measured at the visible wavelengths of 500–800 nm.

3. Results and discussion

Fig. 1 shows the XRD patterns of the p-type films deposited at $R_C = 0$ and 0.5. For the film prepared at $R_C = 0$, the XRD spectrum shows three peaks at 28.3°, 47.2°, and 56.1°, corresponding to the (111), (222), and (311) planes of crystalline Si, respectively. This implies that the film was deposited without introducing CH_4 gas into the plasma,

Table 1

Typical deposition conditions used for preparation of p-SiC:H films.

Parameters	Value
RF power density	0.2 W/cm ²
Base pressure	< 1.33 × 10 ⁻⁴ Pa
Deposition pressure	66.7 Pa
Substrate temperature	150 °C
CH ₄ /SiH ₄ (R_C)	0–2.0
H ₂ /SiH ₄ (R_H)	220
B ₂ H ₆ /SiH ₄ (R_B)	0.4

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