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Growth and characterization of polycrystalline Si films prepared by hot-wire chemical vapor deposition

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Abstract

Hot-wire chemical vapor deposition is a promising method to deposit polycrystalline silicon (poly-Si) at a high deposition rate while maintaining a good material quality. In this paper, the effects of the hydrogen dilution ratio and substrate temperature on the film properties were investigated. Microstructures of the poly-Si films with different deposition parameters have been characterized by Raman scattering, Fourier transform infrared spectroscopy, and transmission electron microscopy. An enhancement in crystallinity was found when the hydrogen dilution ratio and substrate temperature increased. The hydrogen content in the film also decreased when both the hydrogen dilution ratio and substrate temperature increased. Under optimum conditions, the poly-Si film with a grain size of ~0.8 μm and an electron mobility of ~28 cm^2/V s was obtained. These poly-Si thin films have high potential in future low-cost photovoltaic devices.

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1. Introduction

The plasma-enhanced chemical vapor deposition technique has been developed and studied for many years. Hot-wire chemical vapor deposition (HWCVD [1], also called catalytic chemical vapor deposition [2]) is a new technology to obtain device-quality thin films at low substrate temperatures without using a plasma. For the last 10 years, HWCVD has been seriously studied in the laboratory and both the high deposition rate and large area uniformity can be achieved [3,4]. Recently, polycrystalline silicon (poly-Si) and microcrystalline silicon thin films prepared at low temperatures have attracted significant attention because their potential application for low cost, large area electronics to flat panel displays and solar cells [5]. Hydrogenated amorphous silicon (a-Si:H) films can be obtained by HWCVD using pure silane (SiH_4) without hydrogen dilution, which has received considerable attention as an alternative approach for the synthesis of a-Si:H [1,2,6,7]. It has been shown that the hydrogen/silane gas mixture strongly affects the microstructure of silicon films by HWCVD. Hydrogen dilution of silane can lead to a phase transition of silicon from amorphous to microcrystalline and polycrystalline [8,9]. In this paper, the characterization of poly-Si thin films was investigated by Raman scattering, Fourier transform infrared spectrum (FTIR), scanning electron microscopy (SEM), and transmission electron microscopy (TEM). The effects of hydrogen dilution and substrate temperature on the morphology, crystallinity and Si–H bond configuration of the poly-Si thin films are discussed. The electrical

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properties such as Hall mobility and electron concentration will also be described.

2. Experimental details

The poly-Si thin films were deposited using a HWCVD system with a load-lock chamber. The tungsten wire (diameter: 0.5 mm; length: 240 mm) is used as the catalyzer and placed beneath the substrate at a distance of 48 mm. The temperature of the catalyzer is estimated by both an electronic infrared thermometer placed outside a quartz window and from the temperature dependence of the electric resistivity of the catalyzer. The substrate temperature was monitored using a thermocouple attached to the substrate holder. Poly-Si films were deposited on one-side polished (100) Si and glass (Corning 1737) substrates. The substrate temperature ranged from 25 to 450 °C while the filament temperature was kept at 1700 °C. The SiH_4 flow rate was kept at 5 sccm while the H_2 dilution gas was added up to 250 sccm. The deposition pressure was keep at 10 Pa. Before the hot-wire treatment process, the chamber is pumped down to a base pressure of 2×10^{-5} Pa. Details of the deposition conditions are summarized in Table 1.

The thickness and crystallinity of the poly-Si thin film was determined by an alpha-step profilometer (KLA-Tencor P-10) and Raman scattering spectroscopy (Tokyo Instruments Inc.), respectively. The Raman signal due to the transverse optical mode was deconvoluted into three components: I_c (approx. 520 cm^{-1}, crystalline phase with a grain size larger than 10 nm), I_m (approx. 510 cm^{-1}, crystalline phase with a grain size smaller than 10 nm) and I_a (approx. 480 cm^{-1}, amorphous phase) [10]. The crystalline fraction (X_c) was defined as $X_c=(I_c+I_m)/(I_c+I_m+I_a)$. The bonding configurations were investigated by FTIR for samples on (100) Si substrate with a high resistivity. The H content (C_H) was estimated from the Si–H wagging mode (630 cm^{-1}) using a constant of 2.1×10^{19} cm^{-2} [11]. A microstructure factor, R_{IR}, defined as the ratio of the integrated intensity of the 2090 cm^{-1} mode (I_{2090}) to the total integrate intensity ($I_{2000}+I_{2090}$) is used to characterize the structure of a-Si:H/poly-Si thin films. The microstructure of the Si film was also examined by SEM and TEM. Electrical properties of the poly-Si thin films were determined by Van der Pauw–Hall measurements.

Table 1
HWCVD parameters of poly-Si films

Filament temperature	1700 °C
Substrate temperature	25–450 °C
Catalyzer-substrate distance	4 cm
Surface area	754 mm^2
SiH_4 flow rate	5 sccm
H_2 flow rate	0–250 sccm
Gas pressure	10 Pa

3. Results and discussion

The effects of hydrogen dilution ratio and substrate temperature on the Si deposition rate were investigated first. Under a constant silane flow rate (5 sccm), the deposition rate was found to decrease linearly from 0.75 to 0.3 nm/s as the hydrogen/silane gas ratio increased from 0 to 50. Higher hydrogen dilution ratio could result in less available growth radicals and stronger etching processes induced by H atoms, which limited the deposition rate. The substrate temperature seems to have less effect on the deposition rate. The as-deposited Si thin films were analyzed by the Raman spectra to reveal their crystallinity more quantitatively. This approach has been successful in explaining the spectral structure of Raman scattering from small silicon particles [12]. The spectra could be resolved into several components in the region from 400 to 550 cm^{-1}, assuming the Gaussian lineshapes. When the crystalline mode was higher than the amorphous-like one, it separated into two parts, one with a peak at about 520 cm^{-1}, and the other appearing as a broader mode at about 510 cm^{-1}. This separation is probably caused by the distribution of crystallite size (as mentioned above).

The variation in Raman spectra of the as-deposited Si films under different hydrogen dilution ratios is shown in Fig. 1(a). For the Si sample deposited using pure silane, the spectrum shows a peak centered at 480 cm^{-1} with a full width at half maximum (FWHM) of 71 cm^{-1}. This exhibits a typical amorphous feature, where the crystalline Si is calibrated at 520 cm^{-1}. When the H_2/SiH_4 flow ratio increases to 8, the Raman peak shifts to 518 cm^{-1}, indicating the growth of a nanocrystalline Si. That is, the deposition condition is within the transition region from amorphous to microcrystalline phase. It could be due to the reason that the thickness of the amorphous incubation layer became thinner as the H_2 dilution increased. Under a higher H_2 dilution ratio, the amorphous phase Si deposited on the substrate could be etched selectively by hydrogen radicals. In consequence, nucleation of small crystallite occurs directly on the substrate [13]. Fig. 1(b) shows the corresponding X_c and FWHM values as functions of the hydrogen dilution ratio. It was found that the crystalline fraction increased rapidly and then saturated. For the sample deposited with a hydrogen/silane ratio of 50, the X_c value increased to 93% with an FWHM of 7.6 cm^{-1}.

Fig. 2 shows the effect of substrate temperature (T_s) on the X_c and FWHM values of the corresponding Si sample. For $T_s<150$ °C, the film remains amorphous while the film changes from a-Si:H to poly-Si when the T_s increases above 150 °C. It was found that Raman peak shift from 480 to 520 cm^{-1} when the T_s increased from 25 to 450 °C. The corresponding FWHM narrowed from 61 to 7 cm^{-1} and the X_c increased from 12% to 93%. Similar results were also obtained by Han et al. in their HWCVD report [14].

The hydrogen dilution and substrate temperature effects were also investigated by FTIR measurements. In this work,

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