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The formation of poly-Si films on flat glass substrates by flash lamp annealing

Taiki Watanabe, Keisuke Ohdaira *

Japan Advanced Institute of Science and Technology (JAIST), 1-1 Asahidai, Nomi, Ishikawa 923-1292, Japan

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ABSTRACT

We have succeeded the formation of polycrystalline silicon (poly-Si) films by flash lamp annealing (FLA) of 4-μm-thick intrinsic amorphous silicon (a-Si(i)) films deposited directly on flat glass substrates by tuning catalytic chemical vapor deposition conditions. The use of a-Si(i) films deposited without intentional substrate heating leads to the suppression of Si film peeling during FLA. The a-Si(i) films deposited at room temperature have low film density, low film stress, and high defect density, compared to a-Si(i) films deposited at higher temperatures. The prevention of Si film peeling may be due to the low film stress and/or the suppression of the emergence of lateral explosive crystallization by using a-Si(i) films with low film density.

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1. Introduction

In recent years, solar energy has been expected as one of the solutions of environmental problems. Among a variety of solar cells, bulk crystalline silicon (c-Si) solar cells have been widely used and have a market share of more than 80% [1]. However, bulk c-Si cells have some problems such as high cost and using more energy for their fabrication. On the other hand, thin-film Si solar cells using amorphous silicon (a-Si) films deposit by chemical vapor deposition (CVD) can be fabricated with lower costs. a-Si solar cells, however, also have disadvantages such as light-induced degradation and lower conversion efficiency than bulk c-Si solar cells. One of the ways of overcoming these problems is to utilize poly-Si films formed by crystallizing a-Si films [2–4], because they are cost-effective, stable against light soaking, and potentially have high efficiency closer to the efficiency of conventional wafer-based c-Si solar cells of more than 15%. We have so far succeeded the formation of polycrystalline silicon (poly-Si) films by crystallizing precursor a-Si films on glass substrates using flash lamp annealing (FLA) [5–12]. FLA is a millisecond-order annealing technique, and can crystallize μm-order-thick a-Si films without thermal damage to entire glass substrates, because of proper thermal diffusion length on the order of several tens of μm. We have so far clarified that 4.5-μm-thick poly-Si films with a high crystalline fraction can be formed by FLA of precursor a-Si films deposited by catalytic CVD (Cat-CVD) on glass substrates coated with chromium (Cr) adhesion films [5–9]. The advantage of using Cat-CVD a-Si films as precursors is that they contain proper amount of hydrogen and most of the hydrogen atoms remain in poly-Si films formed even after crystallization, which act to terminate dangling bonds in the poly-Si films and contribute to improvement in the quality of the poly-Si films. Although Cr adhesion films significantly contribute to the suppression of Si film peeling during FLA, Cr impurities captured in poly-Si can act as recombination centers and deteriorate solar cell performance. In this study, we have attempted to form poly-Si films, without using Cr adhesion films, from intrinsic a-Si (a-Si(i)) films deposited directly on flat glass substrates by tuning Cat-CVD deposition conditions.

2. Experimental details

We used alkali-free (Corning Eagle XG) flat glass substrates with a size of 19.8 × 19.8 × 0.7 mm^3. After the ultrasonic cleaning of the glass substrates in Semico Clean and isopropyl alcohol, a-Si(i) films with a thickness of 4 μm were deposited on the glass substrates by Cat-CVD at a pressure of 1.1 Pa, catalyzer temperature of 1750 ± 50 °C, a substrate holder temperature from room temperature (R.T.) to 400 °C, and SiH_4 and H_2 flow rates of 50 and 10 sccm, respectively. The typical deposition rate of a-Si(i) films was ~100 nm/min, which was almost independent of substrate temperature. A single shot of 7-ms-duration flash lamp pulse with a fluence of 16 $\mathrm{J/cm}^2$ was supplied for each sample pre-heated at 500 °C in argon (Ar) atmosphere. In order to suppress serious hydrogen desorption from the precursor a-Si(i) films and significant structural variation of a-Si(i) films during the pre-heating, pre-heating duration was limited to be only 3 min. We then evaluated the crystallization of Si films by Raman spectroscopy using the 632.8 nm

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* Corresponding author.
E-mail address: ohdaira@jaist.ac.jp (K. Ohdaira).

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line of a He–Ne laser. The full width at half maximum (FWHM) a c-Si Raman peak obtained from a reference c-Si wafer is ~3.5 cm^{-1}.

We also investigated the characteristics of precursor a-Si(i) films such as hydrogen content, defect density, film density, and film stress. We used Fourier-transform infrared spectroscopy for the evaluation of hydrogen content in a-Si(i) films using samples deposited on high-resistivity c-Si wafers. Defect density in a-Si(i) films was characterized by electron spin resonance by using a-Si(i) films deposited on quartz glass substrates with a size of $2.5 \times 20 \times 0.3$ mm^3. The film density of a-Si(i) films was qualitatively evaluated from their imaginary parts of pseudo-dielectric function obtained by spectroscopic ellipsometry. We characterized the film stress of a-Si(i) films from the bending of Si substrates with a size of $50 \times 7 \times 0.2$ mm^3. About 4-μm-thick a-Si(i) films were deposited on c-Si substrates, and their warping was measured on a stylus profiler. The following Stoney's expression was applied to evaluate film stress [12–14]

$$\sigma = \frac{E_s d_s^2}{6(1-\nu_s) R d_f} \tag{1}$$

where σ, Es, d_s, υ_s, R, and d_f represent the film stress, the Young's modulus of c-Si, the thickness of a c-Si substrate, the Poisson's ratio of c-Si, the radius of curvature, and the thickness of an a-Si film, respectively.

3. Results and discussion

Fig. 1 shows the surfaces of flash-lamp-annealed 4-μm-thick Si films deposited at R.T. and 400 °C. An a-Si(i) film deposited at 400 °C is peeled off after FLA, as shown in the previous report [5]. Other a-Si(i) films deposited with intentional substrate heating show similar results. On the other hand, the a-Si(i) film deposited at R.T. does not peel off even after FLA. Fig. 2 shows the Raman spectrum of the flash-lamp-annealed Si film deposited at R.T. The spectrum of a c-Si wafer is also shown for comparison. A peak located at ~520 cm^{-1}, originating from c-Si phase, is clearly seen in the spectrum of the Si film. This indicates the formation of a poly-Si film by FLA without Si film peeling. The FWHM of the c-Si peak is about 5.6 cm^{-1}, which means that this poly-Si consists of densely packed fine grains with a size of several tens of nm [15]. Since the crystallization of a-Si films through melting process generally results in the formation of much larger crystal grains, the poly-Si obtained is probably formed by solid-phase nucleation and successive growth. These results demonstrate that poly-Si films can be formed with no Si film peeling even without Cr adhesion layers if we carefully tune the deposition condition of precursor a-Si(i) films. c-Si peak in the Raman spectrum of poly-Si film is located at ~519 cm^{-1}, which is lower than that of a reference c-Si wafer of 520.5 cm^{-1}. This fact indicates that the poly-Si film formed has tensile stress. We guess that this is, at least partially, due to the tensile stress of a precursor a-Si(i) film, which is described below, since we have also confirmed the effect of the film stress of a-Si(i) films on that of poly-Si films [8, 10]. It should be noted that there is no evidence of lateral crystallization on the surface of the poly-Si film, as shown in Fig. 1, and the FWHM of the c-Si peak of the Raman spectrum obtained from the flash-lamp-crystallized poly-Si is smaller than that of poly-Si films formed by lateral explosive crystallization (EC) in which solid phase nucleation is dominant and finer grains are likely to be formed [7–9]. This fact indicates that the a-Si(i) film deposited at R.T. is crystallized by a different crystallization mechanism, that is, homogeneous solid-phase nucleation and growth.

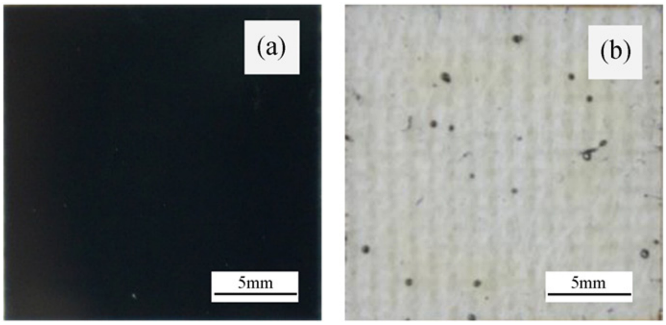

Fig. 1. Surfaces of flash-lamp-annealed 4-μm-thick Si films deposited at (a) R.T. and (b) 400 °C.

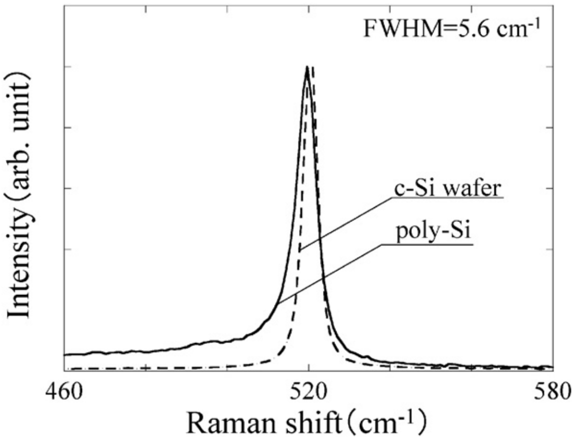

Fig. 2. Raman spectrum of a flash-lamp-annealed Si film deposited at R.T. The spectrum of a c-Si wafer is also shown for comparison.

In order to understand why the peeling of Si films can be suppressed in the case of R.T.-deposited a-Si(i) films, we investigated the properties of the precursor a-Si(i) films. Fig. 3 shows the hydrogen content of precursor a-Si(i) films as a function of deposition temperature. Hydrogen content in a-Si(i) films is an important factor since a higher hydrogen content inside a-Si(i) films generally leads to the peeling of Si films due to the rapid effusion of hydrogen. The hydrogen content of the a-Si(i) films used in this study does not show strong dependence on substrate temperature, and is almost 4% for all the films. This result means that hydrogen content is not a key factor for the suppression of Si film peeling.

Fig. 4 shows the defect density of precursor a-Si(i) films as a function of deposition temperature. An a-Si(i) film deposited at R.T. has a defect density of 3.5×10^{17} cm^{-3}, and defect density decreases monotonically down to ~10^{16} cm^{-3} with increase in deposition temperature. This tendency is generally seen in CVD Si films [16], and can be explained by enhancement in the migration of Si species at higher temperatures. The high defect density might indirectly contribute to the suppression of Si film peeling, although there is no reasonable explanation about

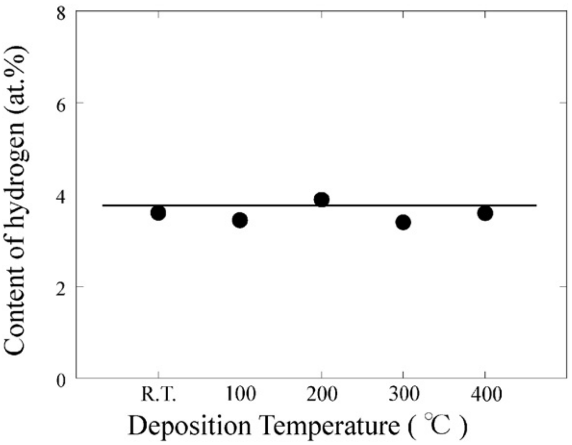

Fig. 3. Hydrogen content in a-Si(i) films as a function of deposition temperature.

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