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Precursor Cat-CVD a-Si films for the formation of high-quality poly-Si films on glass substrates by flash lamp annealing

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ABSTRACT

Amorphous Si (a-Si) films with lower hydrogen contents show better adhesion to glass during flash lamp annealing (FLA). The 2.0 µm-thick a-Si films deposited by plasma-enhanced chemical vapor deposition (PECVD), containing 10% hydrogen, start to peel off even at a lamp irradiance lower than that required for crystallization, whereas a-Si films deposited by catalytic CVD (Cat-CVD) partially adhere even after crystallization. Dehydrogenated Cat-CVD a-Si films show much better adhesion to glass, and are converted to polycrystalline Si (poly-Si) without serious peeling, but are accompanied by the generation of crack-like structures. These facts demonstrate the superiority of as-deposited Cat-CVD a-Si films as a precursor material for micrometer-thick poly-Si formed by FLA.

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1. Introduction

In recent years polycrystalline Si (poly-Si) thin-films formed by post-annealing of amorphous Si (a-Si) have attracted considerable interest as a solar cell material [1-8]. This is because these films show no light-induced degradation due to the absence of a-Si in them: as a result, less Si material is required compared to a bulk crystalline Si solar cell. These advantages result in the fabrication of high-efficiency and low-cost solar cells. However, high temperature processes used to obtain micrometer-thick poly-Si films, which include epitaxial growth on a seed layer consisting of large-grained poly-Si [4,5], hour-order furnace annealing [6,7], and zone melting recrystallization of a-Si films [8], eliminate the possibility of using low-cost substrates. For less thermally resistive substrates such as soda lime glass, we utilize millisecond annealing to sufficiently heat micrometer-thick a-Si films and to avoid thermal damage to glass, which can be realized by flash lamp annealing (FLA). We have already investigated the FLA of 4.5 µmthick a-Si films prepared by catalytic chemical vapor deposition (Cat-CVD), often called hot-wire CVD (HWCVD), which were deposited on glass substrates with the assistance of a Cr adhesion layer inserted between the a-Si and the glass [9], and reported the fabrication of prototype thin-film solar cells using these poly-Si films [10]. Although we attributed the successful crystallization of the Cat-CVD a-Si films, without serious peeling, to their low hydrogen content as well as the

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highly adhesive nature of the Cr films, we were unable to determine which of these factors was more important.

In this paper, we have compared the crystallization, by FLA, of a-Si films deposited both by plasma-enhanced CVD (PECVD) and by Cat-CVD with different hydrogen contents. We have also attempted to crystallize dehydrogenated Cat-CVD a-Si films in order to further understand of effect of the film hydrogen content on the adhesion of a-Si films during FLA.

2. Experimental details

Precursor a-Si films with a hydrogen content of about 3% were deposited by Cat-CVD using deposition conditions described previously [11]. Films that were dehydrogenated were heated at 500 °C under a nitrogen atmosphere for 12 h, resulting in a film hydrogen content less than 1%. We also prepared 2.0 μ m-thick PECVD a-Si films which contained 10% hydrogen; the deposition conditions used were a SiH₄/H₂ flow ratio of 1/5, a chamber pressure of 2 Torr, and a substrate temperature of 200 °C. These Cat-CVD and PECVD a-Si films were deposited directly onto 20 × 20 × 0.7 mm³ quartz substrates to check the adhesion of a-Si without a Cr adhesion layer. The surface roughness of the a-Si films was estimated from atomic force microscopy (AFM) images.

One shot of FLA with a fixed pulse duration of 5 ms was performed on each of the a-Si samples, with its irradiance systematically changed by several tens of J/cm^2 by changing the applied voltage of the lamp. Unfortunately, the actual temperature of the Si films during FLA could not be checked due to the lack of *in-situ* measurement systems. The

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Fig. 1. Surfaces of Si films prepared by Cat- and PECVD after FLA with various irradiance.

lamp-annealed films were characterized by surface photographs, differential interference microscopy, and Raman spectroscopy.

3. Results and discussion

Fig. 1 shows surface images of the lamp-annealed Cat-CVD and PECVD Si films which underwent FLA without dehydrogenation. As can be seen, the 1.5 and 3.0 μ m-thick a-Si films without dehydrogenation are crystallized under proper irradiance, and are accompanied by partial Si peeling. The 3.0 μ m-thick a-Si films are crystallized under a lower lamp irradiance than are the 1.5 μ m-thick films, because the temperature of the a-Si film achieved during FLA is determined by a balance between generated heat and thermal diffusion of this heat into a substrate. From absorption coefficient arguments, thicker a-Si films absorb more flash lamp light, resulting in more heat generation, while the heat diffusion from the a-Si is rather small for the thicker a-Si films due to the difference in thermal diffusion lengths between the a-Si and the quartz. A more detailed description can be found in our previous paper [11].

Unlike the Cat-CVD films, the PECVD a-Si films show a weaker adhesion to the glass substrates. As also seen in Fig. 1, FLA PECVD a-Si films start to peel off even with a lower irradiance than required for crystallization, which never occurs for the Cat-CVD a-Si films. Fig. 2 shows the root-mean-square (RMS) surface roughness of the Cat-CVD and PECVD a-Si films as a function of film thickness. The observed roughness is probably due to the so-called "amorphous-to-amorphous roughening transition", generally observed on surfaces of a-Si films deposited by Cat- and PECVD [12–14]. Although the estimated roughness is large compared to a previous report [15], this might be due to different deposition conditions. The roughness of the PECVD a-Si surface is larger than that observed for the Cat-CVD a-Si roughness, which might be an indication of its weaker adhesion to glass [15].

Fig. 3 shows the surfaces of dehydrogenated Cat-CVD a-Si films after FLA. No significant peeling can be observed after FLA with

relative irradiances of 1.00 and 1.09, under which Si films without dehydrogenation are partially peeled off. This clearly indicates that a-Si films containing less hydrogen show better adhesion to glass substrates during FLA. Clear lateral crystallization features from both edges and spots are seen in the surface photographs of the poly-Si films. A rainbow colored surface, however, does not appear, which is typically observed in the case of films which undergo FLA without dehydrogenation [16]. This might mean that hydrogen atoms in precursor a-Si films are needed for the formation of the 1-µm-pitch grating-like surface structure which enables the observation of the rainbow color. The details of this phenomenon are now under investigation.

Fig. 4 shows the Raman spectra of 3.0 µm-thick poly-Si films formed by FLA, with the same irradiance, both with and without prior a-Si dehydrogenation. The poly-Si films formed using dehydrogenated



Fig. 2. RMS roughness of a-Si films formed by Cat- and PECVD as a function of film thickness. The line is a guide to the eye.

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