

Effect of annealing conditions on polycrystalline silicon produced by the inverted aluminium-induced crystallization of amorphous silicon films on glass substrates

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Abstract. The effect of various annealing treatments on the structure properties of crystalline silicon (c-Si) produced by the inverted aluminium induced crystallization of amorphous silicon (a-Si) films was studied. The surface morphology and grain size of c-Si films were observed by optical microscope and scanning electron microscope. X-ray diffraction and Raman spectroscopy were used to study quantity of Si crystallization due to thermal annealing. Results showed that the c-Si with an average grain size of 54 nm in a (111) orientation was obtained by the thermal annealing at 350 °C for 1 h. Prolonged heat treatment improved Si crystallite quality and increased the average grain size.

1. Introduction

Crystalline silicon (c-Si) films fabricated by a low-cost process are preferable in many semiconductor applications, including thin film transistors and thin film solar cells [1]. There are many different methods to fabricate c-Si films such as solid phase crystallization [2], laser crystallization [3], and chemical vapour deposition. For c-Si films, the fabrication of large grains and high crystalline quality remains the technical challenge due mainly to the fundamental limitations of the thermal budget. Recently, several researchers have used aluminium-induced crystallization (AIC) of amorphous silicon (a-Si) to reduce the thermal budget for producing large grain poly-Si thin films on glass substrate [4-6]. In a conventional AIC, the initial layer structure is glass/aluminum (Al)/amorphous Si (a-Si). Annealing below the Al/Si eutectic temperature (577 °C) results in poly-Si films via a layer exchange process, where the final structure is glass/poly-Si/Al + Si. In the layer exchange process, crystal growth orientation can be vertical and horizontal. The poly-Si is formed in the bottom layer of the films [7].

The conventional AIC must use a chemical etching to remove a layer of Al after the process is finished. On the other hand, an inverted AIC with the initial layer sequence of glass/a-Si/Al has been studied. In this geometry, the final poly-Si is formed on the top surface thus eliminating the necessity for a further lift-off technique. Furthermore, the Al layer underneath the final poly-Si can be directly used as a contact for the solar cell. In this work, we have investigated the influence of the annealing temperature on the formation of the morphological and orientation of the crystallized films prepared by the inverted AIC process. The thickness ratio of Si/Al studied in this work provides large grains and high crystalline quality thin films. These films can be used as a seed layer for preparing a large-grained poly-Si absorber layer for thin film solar cells.

2. Experimental Details

The glass substrates were cleaned in an ultrasonic bath with acetone for 20 min, and were blow-dried prior to layer deposition. Amorphous Si films, with 170 nm thicknesses as measured via cross-sectional scanning electron microscope images, were deposited by RF magnetron sputtering onto glass substrates in the presence of argon gas with a base pressure of 10^{-5} mbar. These Si films were then exposed to ambient air at room temperature for 24 hours before Al deposition to form the silicon dioxide films. The 200 nm thick Al films were subsequently deposited on top of the a-Si layers by RF magnetron sputtering. All the stack layers were then annealed in argon ambient at 250, 300, 350 and 400°C keeping the annealing periods constant at 1 h. After annealing, the samples were cooled slowly to room temperature in the unpowered furnace.

The evolution of the surface morphology of the samples was observed by optical microscope (Nikon SMZ1500). Scanning electron microscope equipped with energy dispersive X-ray (EDX) system (Quanta450, FEI) were employed to investigate the surface morphology, grain size and thickness of the films. X-ray diffraction (XRD) (Bruker D8, Cu K α , $\lambda = 1.541\text{\AA}$) was used to confirm the phase transformation of amorphous to crystalline silicon structure and the films orientation. The crystallinity was determined by Raman spectroscopy (Perkin-Elmer, Spectrum GX system).

3. Results

3.1. Effect of annealing on morphology

The surface variation of films after annealing for 1 h at different temperatures was obtained using an optical microscope and these images are presented in figure 1. At 250 °C, figure 1(a), protrusions having diameters between 5 μm and 50 μm were observed on the film surface. The formation of the protrusions was a stress relaxation mechanism caused by the difference in thermal expansion between Al and Si [8]. The sparse distribution of Si nanocrystals (bright spots) at 300 °C appeared in figure 1(b). The result of figure 1(b) suggested that thermal annealed at 300 °C might not be sufficient for nucleating Si nanocrystals, as shown in figure 1(c) and 1(d).

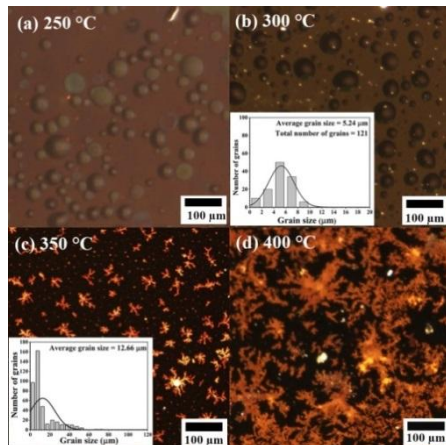


Figure 1. Optical micrographs of samples annealed for 1 h at (a) 250 °C (b) 300 °C (c) 350 °C and (d) 400°C.

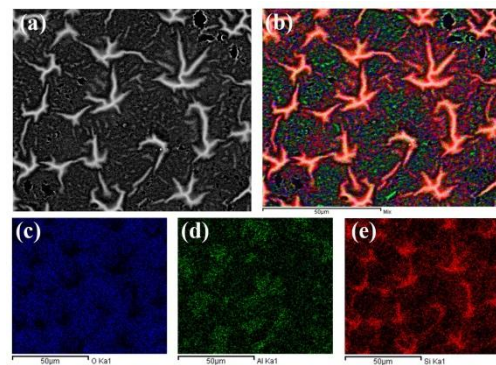


Figure 2. SEM image of the films after annealing at 350°C for 1 h (a), its corresponding EDX mapping (b), distribution of oxygen (c), aluminium (d) and silicon (e) in the sample.

Si nanocrystals, which were confirmed by EDX analysis in figure 2, exhibited dendritic growth at annealing temperatures of 350 °C and 400 °C. The region studied by optical microscope was also investigated by SEM/EDX mapping to identify the surface components. Figure 2(a) is a SEM surface image of the sample after annealing at 350 °C for 1 h. Figure 2(b), (c), (d) and (e) are EDX elemental

mapping components for oxygen, aluminum, and silicon, respectively. The brighter areas corresponded to an abundance of that element. This suggests that Si diffused across the interface layer during the layer exchange process, similar to the results of Duan et al. [9].

3.2. Effect of annealing on structural

XRD spectra of as-deposited Al/a-Si bi-layers film, and after annealing for 1 h at various temperatures are shown in figure 3. Before annealing, no diffraction peak was observed from the as-deposited sample, but clear peaks emerged after the AIC process was completed. Note that after the inverted AIC process, crystalline Si layer was orientated towards the (111) direction. At 250 °C, only a peak of Al indicating a (111) orientation was visible at $\sim 38.6^\circ$.

An onset of Si crystallization whose average diameter was 26 nm appeared after annealing at 300 °C. The crystalline Si layer had (111) orientation at $\sim 28.6^\circ$. At 350 °C, the intensity of Si (111) and Al (111) peaks increased; however, the Al (200) peak was absent. This suggests that the preferential (111) orientation of Si film increased with increasing annealing temperature. This trend holds up to 400 °C, which was the highest temperature used in this study. Interestingly, the intensity of the Si (111) peak from the sample annealed at 400 °C was higher than the Al (111) and Al (200) peaks. A similar result was reported in by Gupta et al. [10]. The Scherrer's equation was applied to determine the mean crystallite size from the XRD results. Peak fitting using a Gaussian function was performed on Si (111) peak. Table 1 summarizes peak locations and estimated c-Si grain sizes for films annealed at different temperatures. As shown in Table 1, the grain size increases with increasing the annealing temperature from 300 °C to 350 °C. However, the grain size decreases with increasing the annealing temperature from 350 °C to 400 °C. The maximum grain size, obtained at an annealing temperature of 350 °C, was 54 nm.

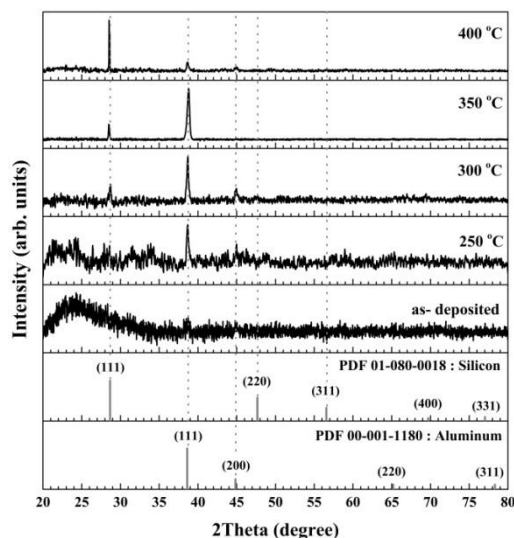


Figure 3. XRD spectra for films annealed at various temperature for 1 h

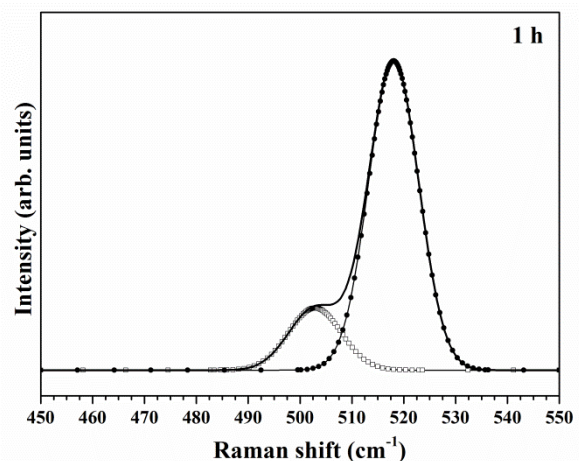


Figure 4. The Raman spectrum for sample annealed at 350°C for 1h.

Raman spectroscopy was used to confirm the crystallinity of the Si films. The Raman spectra were decomposed into amorphous, grain boundary, and crystalline components. The peak intensity of these components occurred around 480, 510 and 520 cm^{-1} , respectively. The Raman peaks in figure 4 were analysed using a peak analysis protocol (Origin 9.1). The films annealed at 350 °C for 1 h showed a peak at 518 cm^{-1} and a peak at 503 cm^{-1} without having an amorphous peak. This observation strongly suggests that the films have a high amount of microcrystalline phase. This is in agreement with the results of x-ray diffraction. The red shifting of Raman peak with full width at half maximum (FWHM)

of about 11 cm^{-1} and the 82.66% crystalline volume fraction observed in these films are due to a change in degree of crystallinity rather than residual stress [11].

Table 1. Summary of structural properties obtained from XRD analyses.

Temperature (°C)	Si(111) Peak Location (degree)	Crystallite size (nm)
250	-	-
300	28.66	26.88
350	28.52	54.20
400	28.66	27.98

4. Conclusions

The formation of single crystal silicon thin films in (111) orientation on glass substrates by inverted AIC method at temperature ranging from 250 °C to 400 °C was demonstrated. The largest grain size of the single c-Si films was observed to be 54 nm. The threshold of the formation of the single c-Si films for this work was the annealing temperature of 350 °C for 1 h. The effects of thermal annealing conditions on the formation of c-Si thin films were studied. The Al/Si exchange process for inverted AIC method was dependent upon the anneal temperature. From X-ray diffraction data, it was found that the Al/Si structural transition from amorphous to crystalline phase after annealing at 350 °C for 1 h. The highest crystalline volume fraction of 82.66% was determined by Raman spectroscopy. These results contribute to the understanding of the Al/Si exchange process and its application to the growth of good quality c-Si thin films on glass substrates.

5. Acknowledgements

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6. References

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