

## Influence of surface roughness of ZnO layer on the growth of polycrystalline Si layer via aluminum-induced layer exchange process

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**Abstract:** This study investigated the effect of surface roughness of zinc oxide (ZnO) layer on the growth of polycrystalline Si layer via an Al-induced layer exchange process. It was found that the growth rate, grain size, crystallization fraction, and preferential orientation of the polycrystalline Si layer were strongly influenced by the surface roughness of the underlying ZnO layer. As the roughness of the ZnO surface increased, a higher growth rate (~40 min) and preferential Si (100) orientation were obtained because of the spatial concentration fluctuations in the Al-Si alloy, induced by the surface roughness of the underlying ZnO layer.

**Keywords:** Solar Cell, Polycrystalline silicon, Zinc Oxide, Surface roughness

### 1. Introduction

In recent years, solar power generation has attracted attention as an important technology in addressing global energy challenges. However, solar power generation is expensive in comparison to conventional power generation methods, because of the high price of solar cell modules. Hence, many studies are underway to realize high quality polycrystalline silicon (poly-Si) films on low-cost substrates. The application of high-quality polycrystalline silicon thin films grown on large-area, low-cost substrates has attracted much attention because of their wide range of applications such as electronics, e.g., for thin-film transistors and solar cells [1][2]. A promising method of preparing a large-area poly-Si layer in a cost-effective manner is the Al-induced layer exchange (ALE) process. The ALE process is a special form of the Al-induced crystallization method, wherein an amorphous Si ( $\alpha$ -Si)/Al/substrate is transformed into an Al/poly-Si/substrate stack by annealing below the eutectic temperature (577 °C) of the Al/Si system [3]-[6]. Because of concentration gradient during annealing, Si atoms diffuse into the Al film through a thin natural aluminum oxide layer that acts as a permeable membrane between the initial Al surface and the  $\alpha$ -Si layer[7]-[9]. As the diffusion of Si atoms continues, a poly-Si layer is formed through nucleation and growth in the Al film. The poly-Si layer

thus obtained can be used as a seed layer for epitaxial growth[2][8]-[11] to produce a thick absorbing layer. The ALE process usually takes several hours, and despite numerous studies, controlling the grain size and primary direction of grains is not yet well understood[12]-[14].

In this study, the use of ZnO for the growth of a poly-Si layer via the ALE method was investigated. The study focused on the effect of ZnO surface roughness on the growth rate, grain size, and orientation of the poly Si layer.

### 2. Experimental

Quartz glass was used as a substrate. It was sequentially washed with acetone, ethanol, and deionized water. A 100 nm thick ZnO film was deposited on the substrate at a rate of 1.6 nm/s via DC magnetron sputtering at room temperature. ZnO layers with varying surface roughness were obtained by controlling the oxygen flow during the deposition.

A 130 nm thick Al layer was deposited on the ZnO layer via thermal evaporation, and it was oxidized in air for 48 h. Finally, an  $\alpha$ -Si layer of approximately 200 nm in thickness was deposited on the Al layer via radio-frequency magnetron sputtering. The deposition rate of  $\alpha$ -Si was approximately 10

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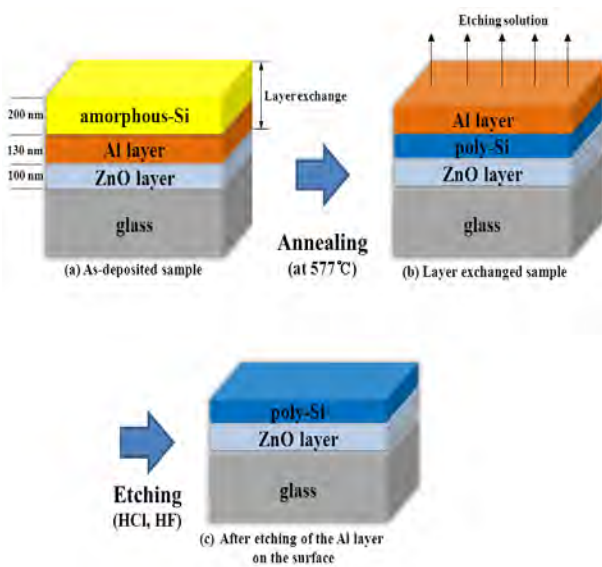
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nm/min, chamber pressure was  $1 \times 10^{-3}$  Torr, and plasma power was 100 W.

As shown in **Figure 1**, the as-fabricated sample has a layered structure:  $\alpha$ -Si/Al/ZnO/glass. The layer-exchange process was performed at 577 °C for 3 h under Ar ambient at a flow rate of 1 L/min. After annealing, the stacking sequence changed into Al/poly-Si/ZnO/glass (**Figure 1(b)**). The annealed samples were taken out of the furnace and the Al layer on the topmost surface was removed using a dilute HCl solution at 40 °C for 5 min, and dilute HF solution for 30 s, at room temperature (**Figure 1(c)**).



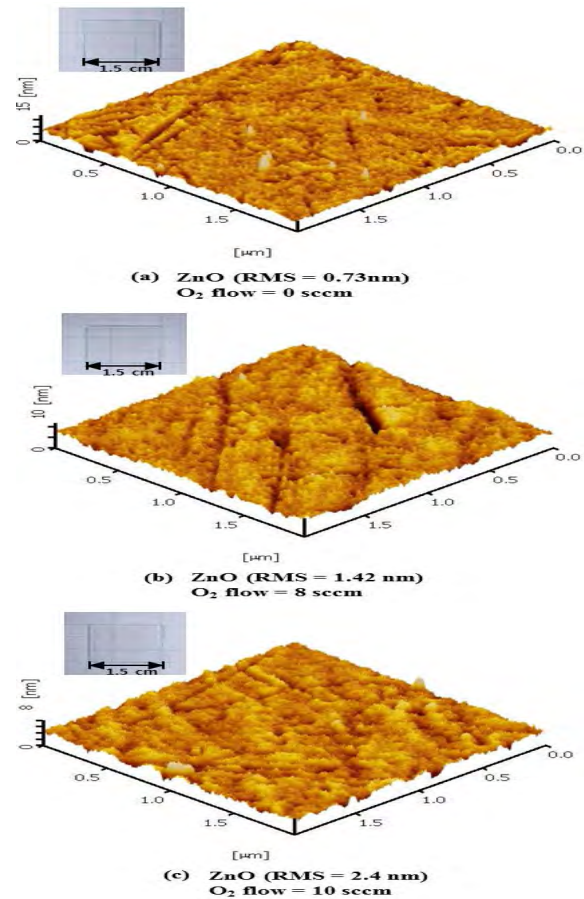
**Figure 1:** Schematic drawing of the fabrication process of poly-Si layer by the ZnO-assisted aluminum-induced layer exchange process: (a) amorphous Si/Al/ZnO/glass stacked sample (as-deposited sample); (b) Al/poly-Si/ZnO/glass stacked sample (layer exchanged sample); (c) poly Si/ZnO/glass (after etching of the Al layer on the surface).

The surface roughness of the ZnO layer was evaluated by atomic force microscopy (AFM, Park systems XE-100). The morphological change of the poly-Si films during annealing was monitored in situ by optical microscopy. The sample surface was observed using Scanning electron microscopy (SEM; JEOL JSM-7000F). The sample surface was polished with a cross section polisher, and it was subjected to cross-sectional SEM analysis. Energy-dispersive spectroscopy (EDS) was conducted to analyze the elemental distribution in the samples. Electron backscatter diffraction (EBSD) was performed to evaluate the orientation of grains; the results

obtained were as follows: the average misorientation angle between the grains was approximately 6° and the electron beam incident angle was 70°, at an acceleration voltage of 20 kV. The beam spot size was 5.4  $\mu$ m and the step size was 0.2  $\mu$ m.

### 3. Results and discussion

**Figure 2** shows the AFM images of the ZnO layer surface. The root mean square (RMS) roughness of the ZnO layer was controlled by changing the oxygen flow during the deposition. The corresponding RMS roughness of the ZnO layer was a) 0.73 nm ( $O_2$  flow = 0 sccm,  $P_{\text{chamber}} = 3 \times 10^{-3}$  Torr); b) 1.42 nm ( $O_2$  flow = 8 sccm,  $P_{\text{chamber}} = 5 \times 10^{-3}$  Torr), and c) 2.4 nm ( $O_2$  flow = 10 sccm,  $P_{\text{chamber}} = 6 \times 10^{-3}$  Torr). As shown in the inset of **Figure 2**, all samples were transparent. Previous studies have shown that an excess of oxygen increases the surface roughness of ZnO layer by restricting the surface migration length[15].



**Figure 2:** AFM images of the ZnO layers grown on glass substrates: (a) ZnO/glass (RMS: 0.73 nm); (b) ZnO/glass (RMS: 1.43 nm); (a) ZnO/glass (RMS: 2.4 nm). The insets in the upper-left corner show pictures of each sample.

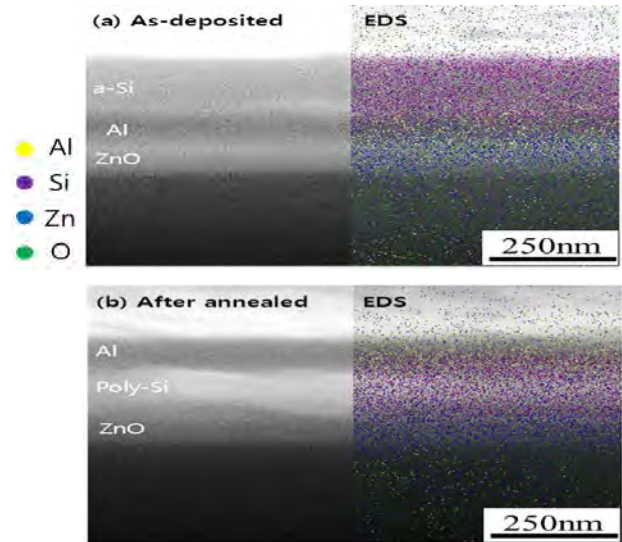
Using the aforementioned ZnO-coated substrates with varying ZnO layer surface roughness, thermal deposition of Al, deposition of  $\alpha$ -Si by sputtering, and the layer exchange process were accomplished sequentially under the same experimental conditions. Three samples were prepared. Samples A, B, and C were prepared on ZnO layers with surface roughnesses of 0.73 nm, 1.42 nm, and 2.4 nm, respectively. The samples were simultaneously annealed at 577 °C.

Schematic of the ALE process is shown in **Figure 3**. It shows the cross-sectional SEM (left-hand side) and corresponding EDS maps (right-hand side) of the as-deposited sample (**Figure 3(a)**) and the annealed sample (**Figure 3(b)**). **Figure 3(a)** shows the  $\alpha$ -Si/Al/ZnO/glass stack, while **Figure 3 (b)** shows an Al/poly-Si/ZnO/glass stack. The layer-exchange process can be explained as follows: at the eutectic temperature of 577 °C, the Si (12.7 %)-Al(87.3 %) alloy begins to melt and Si nuclei form in the Al layer [6][16]. As the melting continues, Si nuclei merge to form a poly-Si layer [11]. The nucleation speed (i.e., growth rate) is affected by various parameters, such as temperature, dissolution flux, and mobility of Si in the Al layer. At a given temperature, the dissolution flux is determined by the concentration of the Al-Si alloy. Ideally, a smooth, abrupt interface between the Al and Si layers will support uniform dissolution flux across the entire interface; a rough interface will induce spatial concentration fluctuations in the dissolution flux.

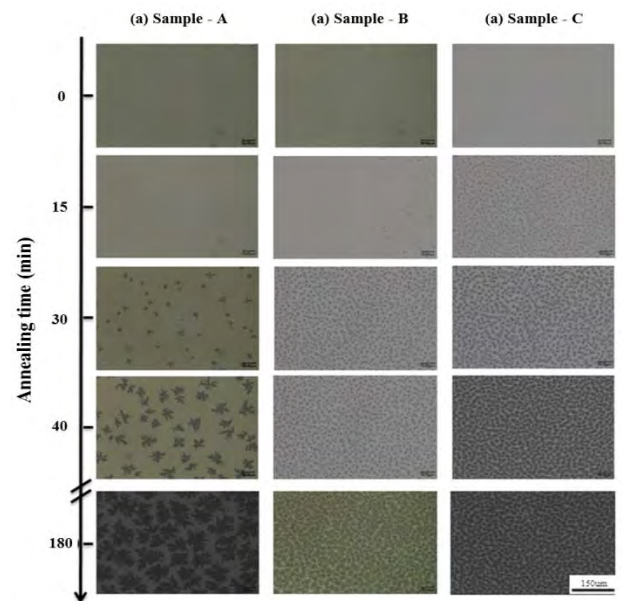
The crystallization process of the poly-Si layer was observed during annealing. **Figure 4** shows optical microscopy images, as seen from the underside, through the quartz substrate. The images of the samples were featureless at the beginning (0 min). In the case of Sample A, low-density, small-size nuclei emerged after 30 min of annealing, and dendritic growth followed, implying a very low growth rate[17]-[20]. In the case of the other two samples, the nuclei emerged much earlier (after ~15 min for Sample B and ~10 min for Sample C), and grew faster as well. The fact that dendritic growth did not occur in the case of Samples B and C indicates an increase in the growth rate. The high growth rate could be attributed to an increase in the amount of the Si dissolution, as the annealing temperature was the same for all samples.

Here it would be appropriate to discuss, in detail, the correlation between the surface roughness of the ZnO layer and the growth rate. There have been several reports on the correlation between the Al-oxide membrane and the growth of Si-nuclei. Klein et al. reported on the increase in growth rate with low quality Al-oxide membranes[21].

Furthermore, Kim *et al.* [9] carried out the ALE process with and without the Al-oxide membrane. With the Al-oxide membrane, they observed a higher growth rate. On the other hand, without the Al-oxide membrane, but a poly Si layer was not obtained. Therefore, a possible explanation for the results in **Figure 4** is that the surface roughness of the ZnO layer affects the dissolution flux of Si by changing the structural quality of the Al-oxide membrane.

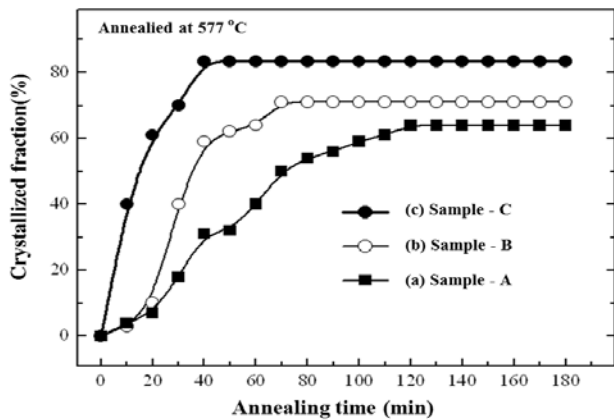


**Figure 3:** Cross-sectional SEM images (left) and EDS mapping images (right): (a) as-deposited sample with a stacking sequence of  $\alpha$ -Si/Al/ZnO/glass and (b) annealed (577 °C) sample, which has a stacking sequence of Al/poly Si/ZnO/glass.



**Figure 4:** Optical micrographs obtained during the layer exchange process: (a) Sample A, (b) Sample B, and (c) Sample C. The black areas in the picture correspond to the poly-Si nuclei.

**Figure 5** shows the variation in the crystallization fraction (CF) as a function of annealing time, showing the growth rate of the Si grains, quantitatively. The CF was quantified from the optical microscope images (**Figure 4**). Sample A revealed ~62% of CF (~20 min), while Sample B showed ~65% of CF (~60 min). The largest CF (~84%), was achieved in the shortest time (~40 min), for Sample C. Note that the reported CF values were in the range of 80–90%, in results of annealed for a few hours or for a few weeks [22]-[24].

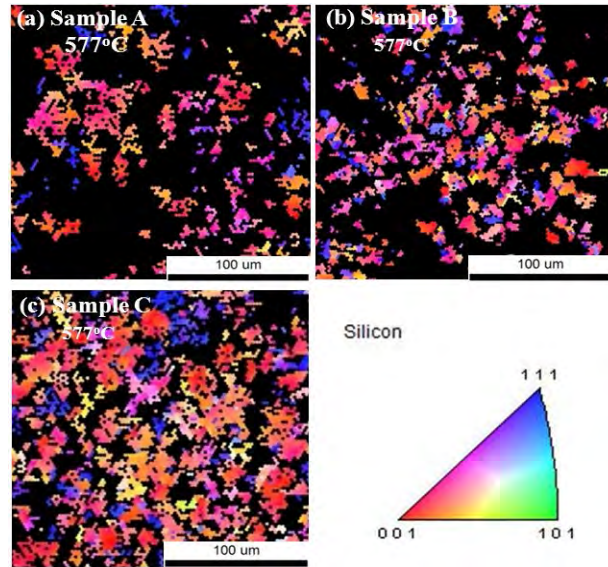


**Figure 5:** Annealing time dependency of the crystallization fraction. The crystallization rate of poly-Si is closely related to the roughness of the ZnO layer.

**Figure 6** shows the in-plane EBSD images. The black area represents  $\alpha$ -Si, and the colored area represents poly-Si clusters. The crystallized area increased with the surface roughness of the ZnO layer. In addition, preferential orientation of the grains is found to be along the Si (100) direction. Note that as the crystallization fraction increases (i.e., as the growth rate increases), more clusters tend to be aligned in the Si (100) direction.

The effect of crystallization conditions, such as substrate and temperature, on the preferential orientation of poly-Si has attracted a lot of attention. In case of dependence on the substrate, a preferential Si (111) orientation was observed on glass substrate [25], while a preferential Si (100) orientation was observed for growth on ZnO surface [26]. Oxidation of the Al layer prior to the deposition of the  $\alpha$ -Si is known to be helpful in obtaining Si (100) orientation. Schneider *et al.* [26] have reported that the nucleation rate of clusters with

an orientation close to the Si (100) direction is higher than that of clusters with other orientations, due to the higher surface energy of the Si (100) plane. Therefore, we conclude that a high growth rate is responsible for the preferential Si (100) orientation of the clusters.



**Figure 6:** In-plane EBSD images of (a) Sample A, (b) Sample B, and (c) Sample C. One can observe that the preferential orientation of poly-Si is closely related to the surface roughness of the ZnO layer.

#### 4. Summary and Conclusion

This study investigated the influence of the surface roughness of the ZnO layer on the ALE growth of poly-Si layer. It was found that both the growth rate and the preferential orientation are closely related to the surface roughness of the underlying ZnO layer. As the ZnO surface roughness increases, a higher growth rate due to spatial fluctuation of the dissolution flux was observed. This induces the preferential Si (100) orientation that is essential for the fabrication of a poly-Si thin film solar cell.

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