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In-situ observation of polycrystalline Si thin films grown using Al-doped ZnO on glass substrate by the aluminum-induced crystallization

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1. Introduction.

Compared to bulk crystalline silicon (including single crystalline and multicrystalline silicon), polycrystalline silicon (poly-Si) thin film can greatly reduce the cost of silicon materials. Recently, the poly-Si thin film grown on glass substrate is attracting for large area electronic and solar cell applications [1,2]. As a common approach of crystallizing amorphous Si (*a*-Si), there are solid phase crystallization [3], excimer laser annealing [4], and metal-induced crystallization (MIC) [5-7]. Among them, the MIC process using metals such as Ni, Ag, Al, and Au is promising to obtain large-grained high quality material with low thermal budget.

To realize poly-Si thin film for high-efficiency solar cells, crystallographic defects, which crucially deteriorate the photovoltaic performance, should be decreased. In order to diminish the crystallographic defects, fabrication of large-grained Si thin film should be required. There are some researches to report on controlling grain size and orientation of poly-Si thin films grown by MIC method [5-7]. They were focused on growth parameters such as metal type, annealing temperature, annealing time, thickness of metal layer, and thickness of metal oxide. Controlling these parameters influences the grain growth velocity especially during initial MIC process, which eventually leads to determination of the grain size and density of poly-Si thin films.

In addition, it is useful if the film could be grown on a textured transparent conductive layer, which can play dual roles for efficient light trapping and transparent electrode. Al-doped ZnO (AZO) is one of the candidates for use as transparent conductive electrodes since it is more resistive to hydrogen plasma and high conductivity [8]. However, its impact on growth kinetics as well as the role of the AZO structure for the AIC process has not been explored.

In this study, we investigate impact of Al-doped ZnO (AZO) layer on Al-induced crystallization (AIC). In-situ monitoring system was utilized to investigate the growth mechanism at growth interface during AIC process. We present structural properties of poly-Si thin films grown with/without AZO layer at different annealing temperature by AIC.

2. Experimental

Glass substrates (2 cm x 2 cm) were cleaned with acetone, methanol, and demonized water. Then, Al layer was deposited on the glass substrate as much as 100 nm by thermal evaporation. Thickness of AZO layer and *a*-Si thin films was controlled as 650 nm and 100 nm by RF sputtering, respectively.

Glass/Al/*a*-Si and glass/AZO/Al/*a*-Si samples were annealed at 500 ^{o}C, 525 ^{o}C, and 550 ^{o}C under argon gas ambient of 1*l*/min during 4 hrs in the in-situ observation system for the crystallization of *a*-Si. The annealing system is shown in Fig. 1. After annealing, the residual Al layer of samples grown with/without AZO was removed by selective wet chemical etching in KOH (5 g KOH: 20 ml H_2O, 0 ^{o}C, 3 min) or HCI (HCl:H_2O=1:2, 40 ^{o}C, 4 min) solution.

The crystallized fraction, grain density, and grain radius of poly-Si thin films layer were investigated by optical microscopy (OM). The crystalline quality of the films was studied by x-ray diffraction and Raman spectroscopy. The orientation mapping of poly-Si films was done using electron backscatter diffraction (EBSD) technique.

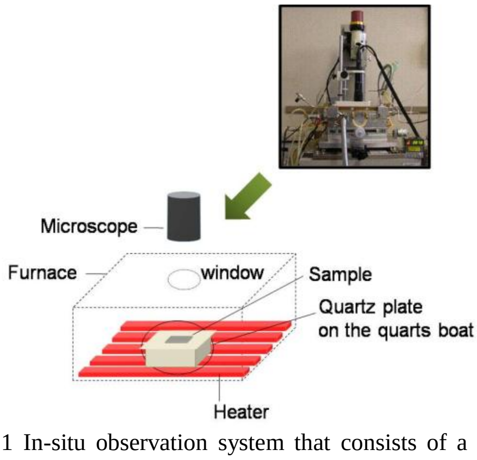

Fig. 1 In-situ observation system that consists of a furnace and a microscope

3. Results

Fig. 2 shows micrographs at the initial glass/Al and glass/AZO/Al interface observed with the optical microscope after various annealing times during an annealing process at 525 ^{o}C. The green and red areas of Fig. 2 correspond to poly-Si grown without/with AZO layer, respectively. In case of the sample grown without/with

AZO, the first Si nucleation was observed after 37 min (Fig. 2(b, upside)) and 25 min (Fig. 2(a, downwards)). Also, the continuous poly-Si film was formed after 62 min (Fig. 2(d, upside) and 54 min (Fig. 2(c, downwards).

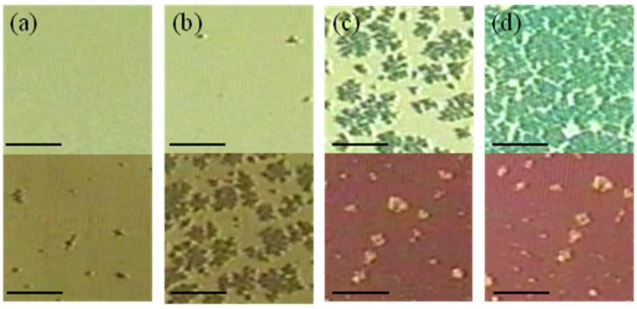

Fig. 2 Optical micrographs of the initial glass/Al (upside) and glass/AZO/Al (downwards) interfaces taken during a crystallization process at 525 ^{o}C. The corresponding annealing times are: (a) 25 min, (b) 37 min, (c) 54 min, and (d) 62 min, respectively. The each scale is 100 µm.

Fig. 3 shows crystallized fraction of the film during AIC as a function of annealing time for different annealing temperature (from 500 to 550 ^{o}C) of samples grown with/without AZO layer. The crystallized fraction of samples grown with AZO was faster than that of samples grown without AZO. Also, as the annealing temperature increases, the average grain size was decreased (Not shown). The decrease of grain size was observed in the samples grown without AZO layer compared to that grown with AZO. The average grain size was increased up to maximum 80 µm at annealed 500 ^{o}C using the glass/AZO/Al (Not shown).

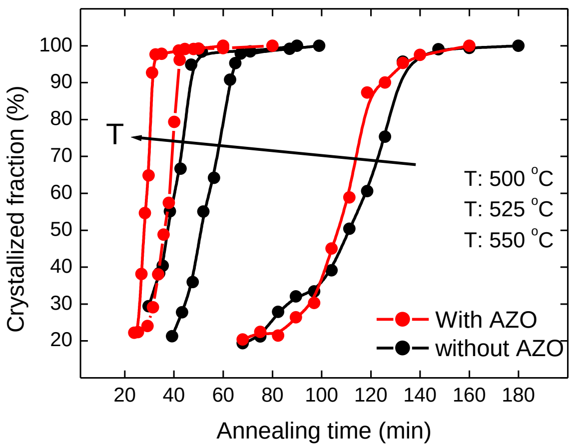

Fig. 3 Crystallized fraction versus annealing time for different annealing temperature of samples grown with/without AZO layer

4. Summary

We studied the crystallization of *a*-Si films in glass/Al/*a*-Si and glass/AZO/Al/*a*-Si during AIC process. The Si crystallization of AZO/Al/*a*-Si during AIC was much faster than that of Al/a-Si. The annealing of glass/Al/*a*-Si led to poly-Si films with small Si grains compared to that of glass/AZO/Al/*a*-Si. At the conference, physics behind the impact of AZO on the growth behavior as well as on preferential orientation and crystallinity of poly-Si films will be discussed.

Acknowledgements

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