



Polycrystalline silicon thin films by aluminum induced crystallization of amorphous silicon

T. Wang^{a,b}, H. Yan^a, M. Zhang^a, X. Song^b, Q. Pan^{b,1}, T. He^b, Z. Hu^b, H. Jia^b, Y. Mai^{b,*}

^a Laboratory of Thin Film Materials, College of Materials Science and Engineering, Beijing University of Technology, Beijing 100124, People's Republic of China

^b Baoding Tianwei Solar films Co., Ltd., Baoding 071051, People's Republic of China

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ABSTRACT

Polycrystalline silicon (Poly-Si) thin films were successfully fabricated on soda-lime glass substrate by aluminum induced crystallization (AIC) process. In order to analyze non-uniform film by AIC, a new method to evaluate the poly-Si thin film average crystalline volume fraction is proposed, based on the optical microscope and Raman spectroscopy results. This method can obtain more accurate crystallization fraction than the common way. X-ray diffraction results showed that the films are strongly (1 1 1) orientated. A new region crystallization pattern in AIC was also proposed.

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1. Introduction

One of the most challenging issues for the development of polycrystalline silicon (poly-Si) thin-film solar cells is the growth of crystalline silicon on low-cost substrates at low temperature. An approved way to overcome this problem is the deposition of amorphous silicon (a-Si) with subsequent low-temperature crystallization process. Among those processes is solid phase crystallization (SPC) [1], laser induced crystallization (LIC) [2], and metal induced crystallization (MIC) [3,4]. MIC is considered as the most competitive way for industrial manufacturing, owing to the mature annealing process at a low temperature, which may strongly reduce the production cost as compared to the high temperature process in the conventional crystalline silicon solar cell fabrication. The aluminum induced crystallization (AIC) is one type of MIC, whose crystallization temperature was found to be dependent on the thickness ratio of aluminum (Al) over silicon (Si) [5]. Until now, a temperature as low as 150 °C has been reported [6]. However, poly-Si films prepared from AIC process were heavily doped with Al [7] and thickness limited [8]. Thus they are suitable for the p⁺ layer of the poly-Si thin film solar cells or the seed layer for the poly-Si absorber layers [9–11].

Although many works about the AIC process have been reported before [12–14], the growth process still needs further study, such as

the phenomenon of region crystallization. Widenborg and Aberle [15] investigated the surface morphology of poly-Si thin film during AIC process and built a schematic model to explain the aluminum-induced layer exchange (ALILE) process. However, the detailed analysis about growth of grain has not been performed yet, due to the difficulties in differentiating grains in optical microscope (OM) and scanning electron microscope (SEM). In this paper, we chose a simple method, by controlling the Al/Si thickness ratio, to obtain an easily analyzable poly-Si thin film. Meanwhile, a new way to evaluate the film crystallization fraction is introduced. OM, Raman spectra and X-ray diffraction (XRD) are used to characterize the film crystallinity.

2. Experiment

In this study soda-lime glass was used as substrate for Al and a-Si deposition. Prior to the deposition, the glass substrates were first ultrasonically cleaned for 10 min with deionized water and detergent TDA7 from Franklab, and then dried with high pressure N₂ purge. A 300 nm thick aluminum film was then sputtered on glass in Ar gas at room temperature and then oxidized in atmosphere naturally for 3 weeks. An amorphous oxide thin film (alumina) with reasonable thickness homogeneity is formed on Al layer during the oxidation process [16]. With increasing alumina thickness, alumina would passivate the surface and hinder further oxidation. An oxidation of 3 weeks was long enough to reach a saturation of alumina thickness which was found to be less than 5 nm at room temperature [17]. Thus just top surface is oxidized and acts as a diffusion barrier layer in AIC process. Subsequently, a-Si film was deposited on the Al film by plasma enhanced chemical vapor

* Corresponding author. Tel.: +86 312 3309996; fax: +86 312 3309866.

E-mail addresses: yfcx@btw-solarfilms.com, qtpan00@hotmail.com (Q. Pan), y.mai@btw-solarfilms.com (Y. Mai).

¹ Tel.: +86 312 3309927; fax: +86 312 3309866.

deposition (PECVD) in a multi-chamber cluster tool from MVSys-tem. The base pressure and working pressure for a-Si deposition were kept at 7×10^{-4} Pa and 65 Pa, respectively. Silane (SiH_4) flow over hydrogen (H_2) flow ratio (SiH_4/H_2) was 1:5, and total gas flow was 36 sccm. The thickness of a-Si film was around 250 nm.

The as-deposited samples were annealed at 460 °C and 500 °C in dry nitrogen ambient for different durations ranging from 10 to 360 min. To exclude the effect of cooling rate of different annealing temperatures on total annealing times, all samples were quickly unloaded from the oven and cooled down to room temperature (RT) in atmosphere within 5 min. After the annealing, part or most of the Al layer would be exchanged from the bottom to the top of the Si layer. The Al matrix was selectively removed by a standard etching procedure [18] (80% of phosphoric acid, 5% of nitric acid, 5% of acetic acid and 10% of deionized water at 50–55 °C for 20 min), and the Si layer will be exposed after the etching.

Optical microscope (OM, Olympus BX51M) was used to investigate the surface morphology of the samples. Raman spectroscopy

(HORIBA, T6400) and X-ray diffraction (XRD, Bruker D8 Advance) were used to determine the crystalline fractions (CF) and crystal orientation, respectively. In our study, all Raman scattering measurements were done with a focused green laser beam ($\lambda = 514.5$ nm) at a beam power of 4.9 mW to prevent laser induced crystallization. A 50 \times long distance objective lens was used to form a laser spot size of $2 \mu\text{m}^2$ on the sample. Thus, the Raman spectra of individual micro-region can be handily obtained. Such laser has a penetration depth $1/\alpha$ of about $1 \mu\text{m}$ in silicon [19].

The surface of poly-Si samples after the annealing and etching process consists of different zones, such as grain, mesh structure, etc. The areas of different zones were obtained by analyzing the OM image of the samples using imaging tools, such as Image-Pro Plus 6.0 and Photoshop CS5. OM images were taken of randomly selected regions on the 10 mm \times 10 mm poly-Si samples. The total pixels of each image is 2560 px \times 1920 px, corresponding to 0.02 mm² on the surface of the poly-Si samples. The grain and mesh structure regions can be isolated and recognized automatically by the imaging tools.

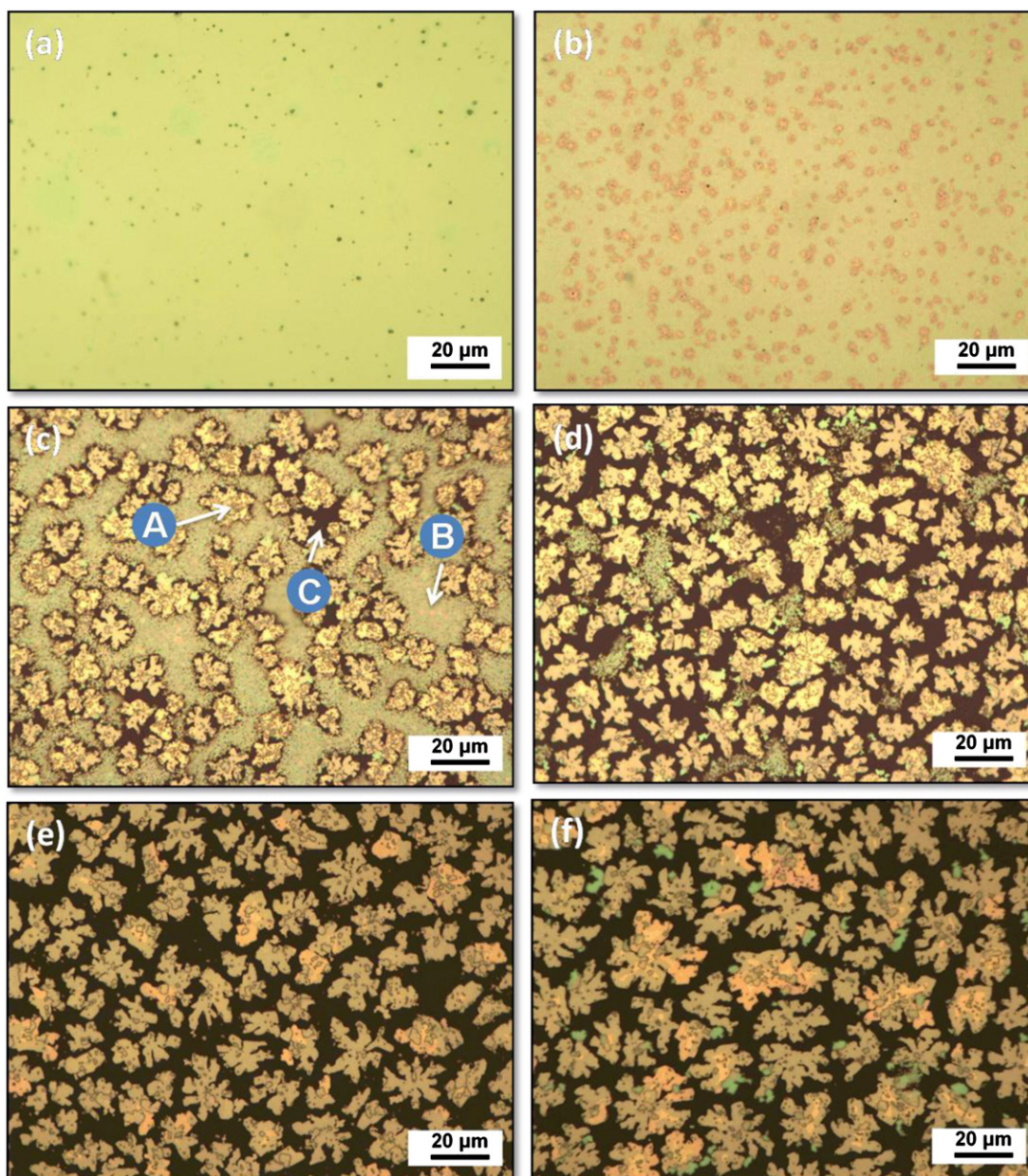


Fig. 1. Optical micrograph of samples annealed at 460 °C for (a) 20 min, (b) 30 min, (c) 60 min, (d) 90 min, (e) 150 min and (f) 240 min.

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