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Structural properties of polycrystalline silicon films formed by pulsed rapid thermal processing*

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A novel pulsed rapid thermal processing (PRTP) method has been used for realizing solid-phase crystallization of amorphous silicon films prepared by plasma-enhanced chemical vapour deposition. The microstructure and surface morphology of the crystallized films were investigated using x-ray diffraction and atomic force microscopy. The results indicate that PRTP is a suitable post-crystallization technique for fabricating large-area polycrystalline silicon films with good structural quality, such as large grain size, small lattice microstrain and smooth surface morphology on low-cost glass substrates.

Keywords: polycrystalline silicon film, rapid thermal processing, microstructure
PACC: 6110M, 6170A, 6140, 8139H

1. Introduction

Polycrystalline silicon (poly-Si) is a material of great importance in microelectronics, photovoltaics and display technologies.[1−5] In recent years, there has been growing interest in developing practical techniques for fabricating large-area and high-quality poly-Si films on low-cost glass substrates at low temperatures. Generally, post-crystallization of amorphous silicon is thought to be a preferred technique for this purpose.[6−8] Compared to those directly-grown, the poly-Si films prepared by post-crystallization have a perfect structure and smooth surface morphology that are required for critical applications. Several post-crystallization methods have been developed, among which zone-melt recrystallization (ZMR) can produce large-grain poly-Si films,[9] but the very high process temperature ($> 1000^\circ$C) makes the use of low-cost substrates actually impossible. Conventional furnace annealing requires several tens of hours at temperatures around 600°C (the softening point of glass),[10,11] which means high thermal budget and low throughput, uneconomical for large-scale production. Rapid thermal processing (RTP) can greatly shorten the process time,[12,13] but due to the continuous thermal exposure to high temperature, the damage to the glass substrate is inevitable. Although laser beam annealing can realize the crystallization at lower temperatures,[14,15] it is very expensive and also needs to take a long time to scan over a large area. Post-crystallization using a metal as seed can also be carried out at temperatures lower than 600°C,[16−18] but the contamination of metal impurity to the Si film is induced. Therefore, developing a new effective method to prepare poly-Si films on low-cost substrate is still an attractive and challenging target.

Currently, we have proposed an improved rapid thermal processing method—pulsed rapid thermal processing (PRTP)—which involves a number of cycles of thermal process. In each cycle, a 60s thermal bias of 550°C is used for pre-heating the films to a "critical state" of crystallization so that the rapid nucleation and grain growth can be triggered by a following 1s high-temperature thermal pulse of 850°C. By using this annealing method, solid-phase crystallization in amorphous silicon films can be completed in several minutes, without the need for a metal seed. Moreover, since the thermal bias temperature of 550°C is well below the softening point of Corning 7059 glass and the samples are exposed to the high-temperature pulse of 850°C for a very short time (only 1s) in each cycle, the thermal damage to the glass sub-

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strate can be effectively minimized.

Many of the electrical properties of poly-Si depend upon its structural quality since carrier transport is sensitive to the lattice imperfection. Perfect structure and smooth surface are desirable for device-quality poly-Si films. In this paper, we investigate the microstructure and surface morphology of the PRTP-prepared poly-Si films by using x-ray diffraction (XRD) and atomic force microscopy (AFM). The structural properties of the poly-Si films are characterized in terms of crystal orientation, grain size, lattice microstrain and surface roughness. The effect of the deposition parameters of the precursor amorphous silicon on these structural qualities of the crystallized films is also discussed.

2. Experimental details

The precursor amorphous silicon films for post-crystallization were prepared by the plasma-enhanced chemical vapour deposition (PECVD) method at different deposition rates by changing the ratio of gas flow (SiH_4/H_2). Typical deposition parameters are summarized in Table 1. The material used for substrates was Corning 7059 glass. The film thicknesses were determined by the optical transmission spectra, and the deposition rates (r_d) were calculated by dividing the measured film thickness by the deposition time.

Table 1. Deposition parameters of amorphous silicon films.

Sample	Substrate temperature/°C	H_2/SiH_4	Time/h	Thickness/μm	r_d/(nm/s)
S1	400	10	4	0.932	0.0647
S2	400	8	4	1.213	0.0842
S3	400	5	4	1.435	0.0995
S4	400	3	4	1.773	0.1231

The crystallization by means of PRTP was carried out in a KST-2 rapid thermal processor with thirteen tungsten halogen lamps as heat source and N_2/Ar as ambient atmosphere. The heating rate is about 300°C/s. Samples for treatment were placed on a silicon wafer holder, on which besides the samples a Pt–PtRh thermocouple was attached, which was used for measuring and controlling process temperature. The programmed and actual time–temperature patterns of the PRTP are shown in Fig.1.

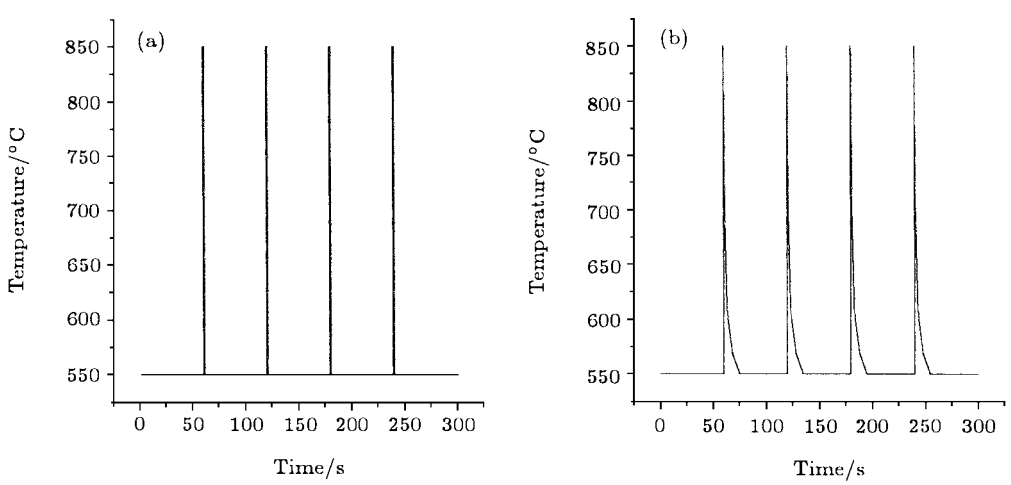


Fig.1. The programmed (a) and actual (b) time–temperature patterns of PRTP.

X-ray measurements were performed using a Philips diffractometer, and the diffraction profiles were recorded by automatic Savitzky–Golay scanning with Ni-filtered Cu$K\alpha$ radiation (0.1542nm). With accurate measurements and strict analysis of the diffraction peak, it is possible to achieve a reliable

analysis of the lattice microstrain and a reasonable evaluation of the mean grain size which was usually overestimated by averaging the sizes of the maximum grains observed in the TEM images. AFM analyses of both annealed and unannealed films were performed using a Digital Instrument Nanoscopy III multimode microscopy, operating in a tapping mode.

3. Results and discussion

Figure 2 shows the x-ray diffraction profiles of sample 4 on glass before and after 1-, 5-, 10- and 20-cycle PRTP respectively. No obvious diffraction peak was observed before PRTP, exhibiting that the as-deposited Si film (a) is structurally amorphous. Several small peaks appear after 1-cycle PRTP (b), indicating the initiation of crystallization. Then the peak intensities are enhanced with increasing process time and reach stable values after 10-cycle processing, showing that the post-crystallization can be completed within several minutes, which is two orders of magnitude shorter than the time required by the conventional furnace annealing method. We can see the very strong diffraction peak at 28.52° that represents (111) preferred orientation. In general, the crystallite size can be evaluated from the half-peak width of the x-ray diffraction peak using the Scherrer formula $D(hkl) = k\lambda/[\beta(2\theta)\cos\theta]$. This formula can give a mean grain size assuming that the peak line broadening only results from the size distribution after subtracting the instrument broadening from the measured full width at half maximum (FWHM) $\beta(2\theta)$. Actually, with an increase of crystallite size the peak broadening due to lattice imperfections such as microstrains becomes considerable. To obtain reliable values of the crystallite size in this case, we used the Fourier analysis method[19] that was originally used to evaluate the structural quality of cold-worked metals. This method is based on the analysis of the peak profile rather than the estimation of the half-peak width. Following the work of Warren and Averbach,[20,21] the diffraction profile was fitted by a Fourier series after subtracting the background and $K\alpha_2$-component profile by making the Rachinger correction.[22] Then the mean grain size and root-mean-square (rms) strain can be determined from a plot of the natural logarithm of the Fourier cosine coefficient versus the square Miller index sum $(h^2+k^2+l^2)$.

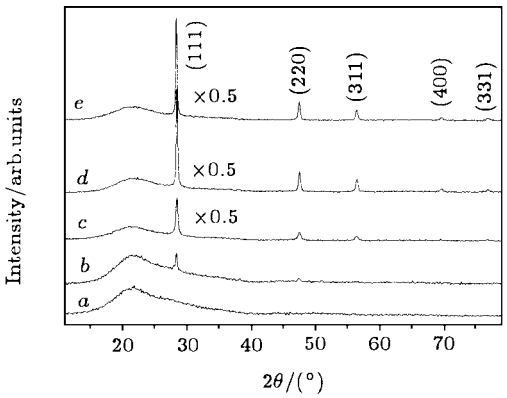


Fig.2. XRD patterns of a Si film (S4) before PRTP (a) and after 1-cycle (b), 5-cycle (c), 10-cycle (d), 20-cycle (e) PRTP.

To study the effect of deposition parameters on the crystallization, the four samples (S1–S4) deposited at different rates were processed by PRTP for 10min. The crystallization was then completed and the stable values of grain size and microstrain were obtained by XRD as mentioned above. Figure 3 shows the grain size and microstrain as functions of deposition rate of the precursor amorphous films. It can be found that higher deposition rates result in larger grain sizes and smaller microstrains. This can be explained as follows. With an increasing deposition rate, the structural disorder of the Si network increases, which usually results in low nucleation rates[11] during the post-crystallization procedure, thus giving rise to larger grain sizes and smaller lattice strains.

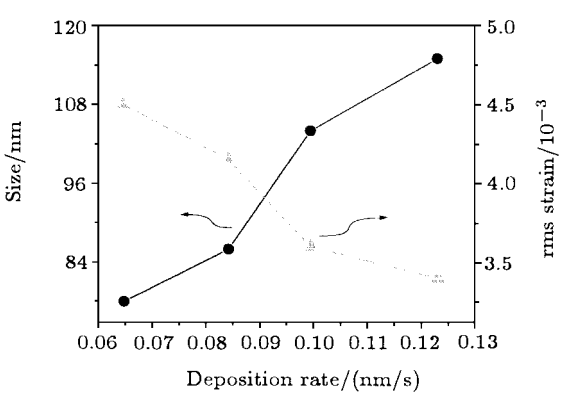


Fig.3. Grain size and microstrain as functions of deposition rate of the precursor amorphous films.

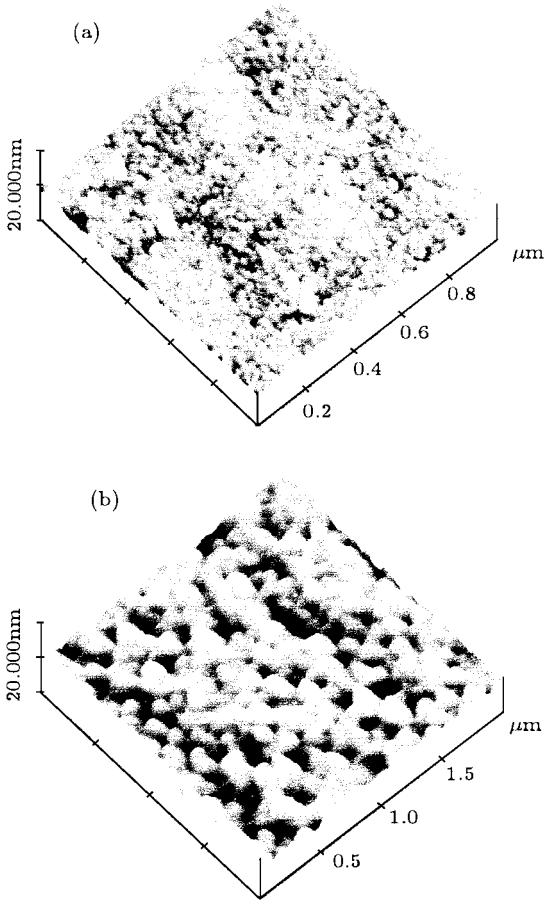


Fig.4. AFM images of a Si film (S3) before (a) and after (b) PRTP.

Figure 4 shows AFM images of sample 3 before and after 10-cycle PRTP. The image (b) reveals the surface morphology of the crystallized film which correlates well with the grain sizes evaluated from XRD profiles. Furthermore, rms roughness was calculated from "raw" $1\times1\mu m^2$ scans and transformed into k-space with the k_x=0 and k_y=0 lines removed to correct for AFM artifacts.[23] As a result, the rms roughness of sample 3 before and after PRTP is found to be 0.580 and 0.955nm, respectively. Such a roughness of the crystallized film is one order of magnitude smaller than that[24,25] of the directly-deposited poly-Si film with the same thickness.

4. Summary

Using XRD and AFM, we have examined the microstructure and surface morphology of poly-Si films formed by pulsed rapid thermal processing. This newly-developed post-crystallization technique has the potential for preparing poly-Si films with large grains and smooth surface. Moreover, the mean grain size and microstrain in these poly-Si films are closely related to the deposition parameters of the precursor amorphous silicon films. A higher deposition rate results in larger grain size and smaller lattice strain in the crystallized films.

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