JOURNAL OF MATERIALS SCIENCE 28 (1993) 6279–6284

A study of the morphology and microstructure of LPCVD polysilicon

EUN GU LEE
Department of Materials Engineering, Chosun University, 375, Seosuk-Dong, Dong-Gu, Kwangju 501-759, Korea

SA KYUN RHA
Semiconductor Process Division (8th Laboratory), Goldstar Electron Co. Ltd, 16, Woomyeon-Dong, Seocho-Gu, Seoul 137-140, Korea

The morphology and microstructure of polysilicon films deposited by low-pressure chemical vapour deposition (LPCVD) have been investigated as a function of deposition conditions. The deposition temperature was varied from 540–640 °C. As-deposited polysilicon films had a rough surface with (1 1 0) textured columnar grain structure, while as-deposited amorphous films had a smooth surface. The polysilicon film deposited at the amorphous to polycrystalline transition temperature had an extra-rough, rugged surface with (3 1 1) texture. At the transition temperature, the grain structure tended to shift from the polycrystalline to the amorphous state with increasing deposition pressure and film thickness. It was found that nucleation of amorphous film during *in situ* annealing at the transition temperature without breaking the vacuum began to occur from surface silicon atom migration, in contrast to a heterogeneous nucleation during film deposition.

1. Introduction

Polycrystalline silicon (polysilicon) films formed by low-pressure chemical vapour deposition (LPCVD) of silane (SiH_4) are widely used in integrated circuits for various applications as MOS gates, interconnects, resistors, and emitter contacts. Other applications include photovoltaic conversion, thermal and mechanical sensors, and thin film transistors (TFT) for large-area liquid-crystal displays (LCDs). The electrical performance of the polysilicon is strongly determined by its microstructure, which depends on deposition parameters [1–4]. Electrical properties for as-deposited polycrystalline or as-deposited amorphous silicon have been investigated by a number of authors [5–7]. It was suggested that deposition temperature should be as low as possible to obtain high conductivity and carrier mobility.

It is well known that the surface roughness of polysilicon degrades the electrical characteristics of dielectric film on the polysilicon [6, 8, 9]. However, in order to obtain the sufficient storage capacitance required for 64M bit dynamic random access memory (DRAM) and beyond, some fabrication technologies having an uneven surface of hemispherically grained (HSG) polysilicon film have been suggested for increasing effective surface area [10, 11].

In the present work, we investigated the deposition condition dependence of surface morphology and microstructure of LPCVD polysilicon. The grain growth mechanism during the deposition and subsequent *in situ* annealing at the amorphous to crystalline transition temperature without breaking the vacuum has been also discussed.

2. Experimental procedure

The experiments were carried out in an induction-heated hot-wall horizontal reactor. Undiluted monosilane (SiH_4) gas as a silicon source was supplied from both sides of the tube and was evacuated using a rotary pump. The deposition parameter, temperature and pressure, were varied within the constraints of equipment available and film quality. The starting wafers were CZ(1 0 0) p-type silicon, with 100 nm thick silicon dioxide (SiO_2). The film thickness was measured using an ellipsometer, and was approximately 100 nm unless otherwise specified. All films were undoped and were visually inspected with an oblique light and a qualitative assessment of film quality based on the hazy spot.

The preferred orientation (texture) of the films was investigated using an X-ray diffractometer (XRD) with a glancing incident angle to reduce the penetration depth of X-rays and hence the diffraction from the substrate silicon. The texture of the films was measured by comparing the intensity of diffraction peaks with those obtained on randomly oriented polycrystalline film. In order to quantify the texture of the films, the relative unit ($r.u_{hkl}$) for each diffraction plane (hkl) is normalized to the (1 1 1) plane as follows.

$$u.r_{hkl} = I_{hkl}/I_{111} \quad (1)$$

0022–2461

where $I_{h\,k\,l}$ and $I_{1\,1\,1}$ are intensities of $(h\,k\,l)$ and (1 1 1) planes, respectively. Film morphology and microstructure were investigated using plane and cross-sectional scanning electron microscopy (SEM) and transmission electron microscopy (TEM).

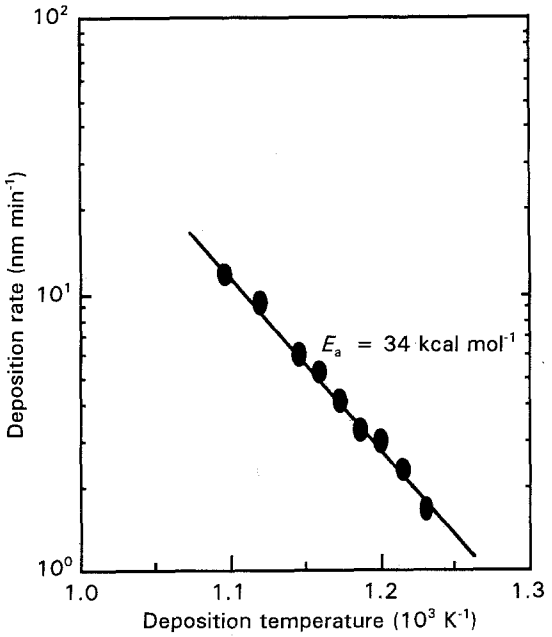


Figure 1 Arrhenius plot of the deposition rate for 0.25 torr silane pressure.

3. Results and discussion

Fig. 1 shows the Arrhenius plots of growth rates in the temperature range 540–640 °C under 0.25 torr. The silane growth is controlled by surface reaction and has an apparent activation energy of 34 kcal mol^{-1}, which agrees well with 32–39.9 kcal mol^{-1} reported by Harbeke *et al.* [4]. The surface morphology of the films at various deposition temperatures is shown in Fig. 2. It was observed that the film deposited at 560 °C has a smooth surface and the film deposited at 570 °C has some nuclei, which start to grow in the amorphous phase. The film deposited at 580 °C has hemispherical grains (HSG), while at 590–600 °C it has an extra-rough, rugged surface with a greater surface profile variation, and at 620 °C has a rough surface. The surface morphologies of the specimens annealed at 1000 °C for 4 h were similar to those of as-deposited specimens. These facts imply that the surface morphology of the film is strongly dependent on the deposition temperature, but is almost independent of annealing conditions.

Crystal structures of the films were analysed using XRD. Three X-ray diffraction peaks showing (1 1 1), (1 1 0), and (3 1 1) reflections were detected for the samples deposited at and above 570 °C. Fig. 3 shows the ratios of (1 1 0) and (3 1 1) intensity to (1 1 1) intensity as a function of deposition temperature. For a

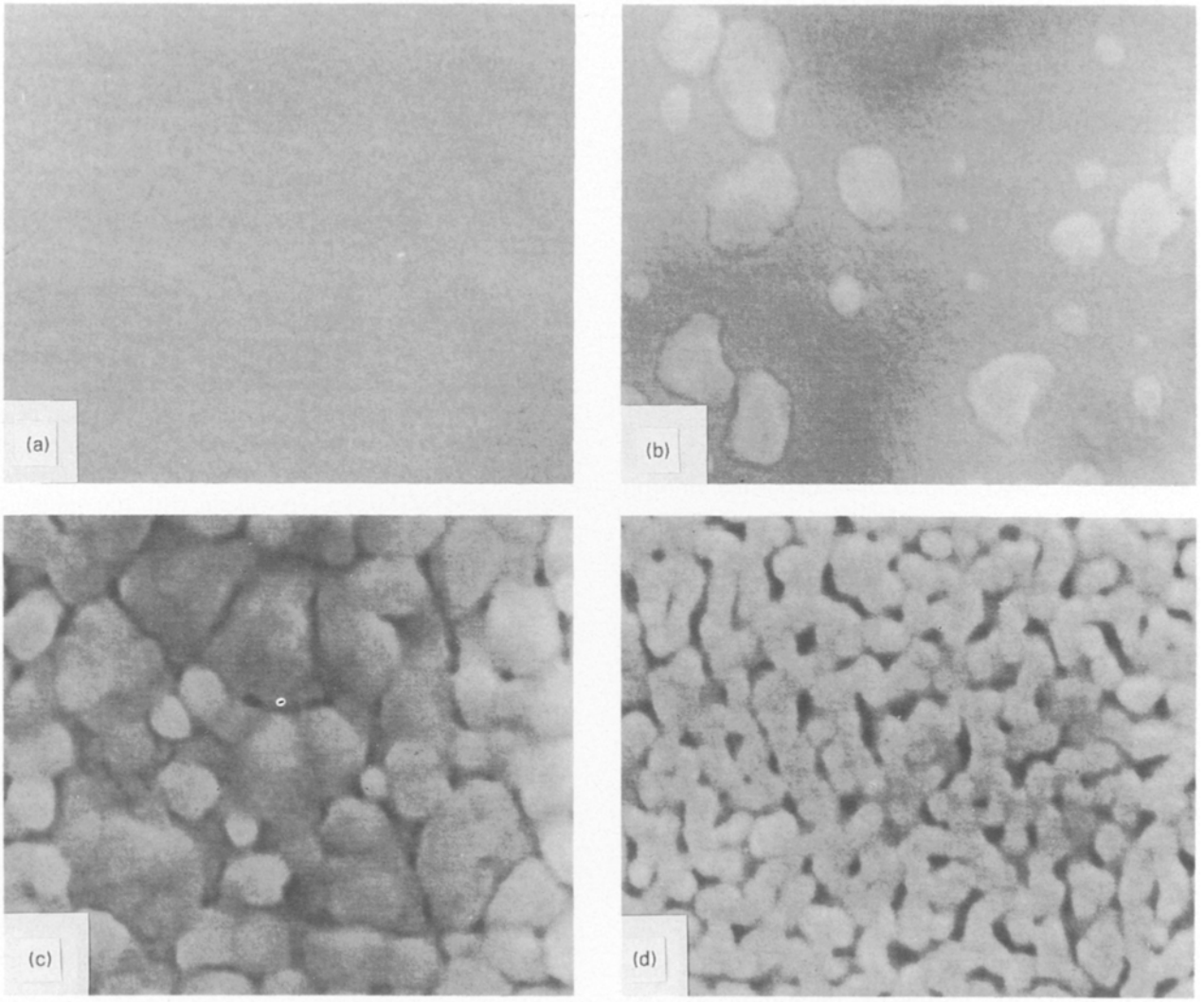


Figure 2 Scanning electron micrographs of films deposited at (a) 560 °C, (b) 570 °C, (c) 580 °C, (d) 590 °C, (e) 600 °C, and (f) 620 °C.

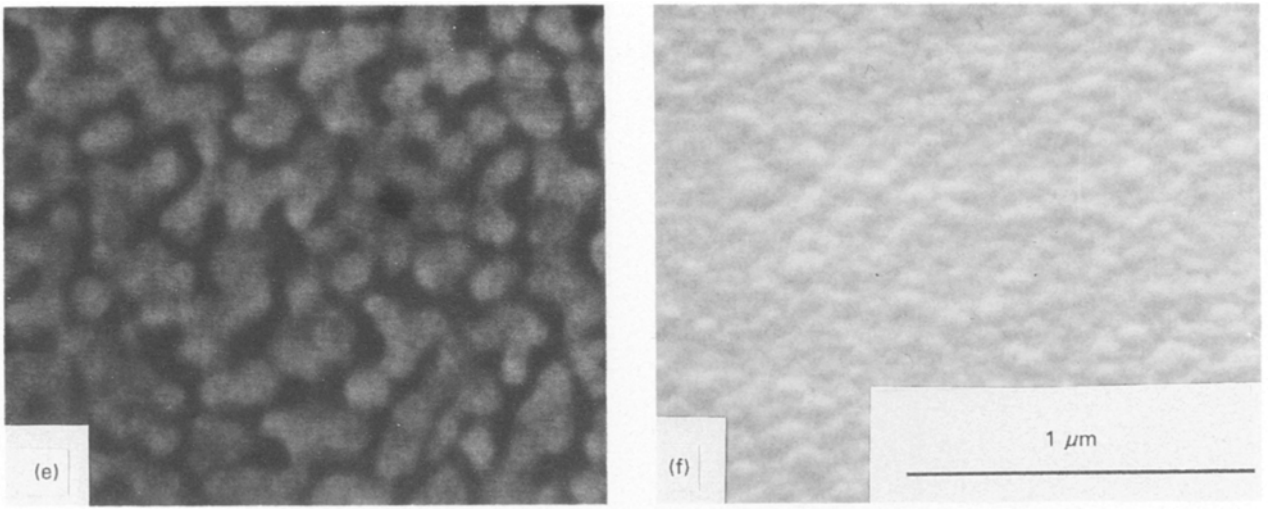


Figure 2 Contd.

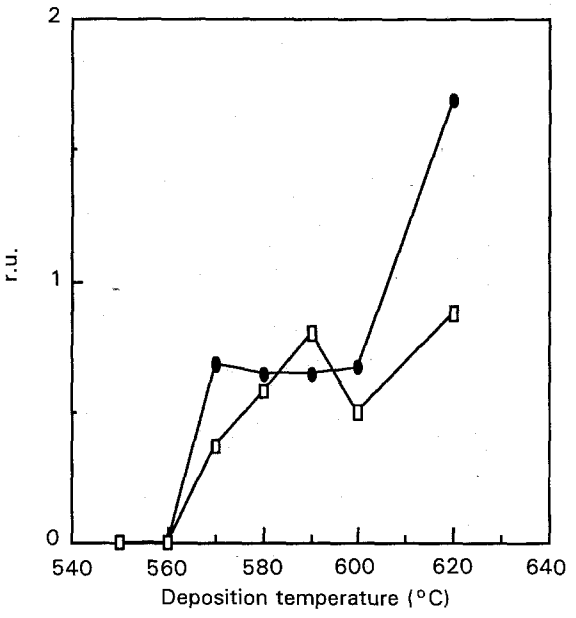


Figure 3 Relative unit of (●) (1 1 0) and (□) (3 1 1) intensity to (1 1 1) intensity as a function of deposition temperature.

randomly oriented polysilicon film, the values of $I_{1\,1\,0}/I_{1\,1\,1}$ and $I_{3\,1\,1}/I_{1\,1\,1}$ are 0.6 and 0.35, respectively. The film deposited below 560 °C is amorphous, because no XRD peaks were detected. It was observed that the (3 1 1) component dominates for the films deposited in the temperature range 580–600 °C and the (1 1 0) component is a major peak for the films deposited above 620 °C. The dramatic increase in (1 1 0) texture above 620 °C indicates that the preference for (1 1 0) texture is a growth phenomenon rather than nucleation behaviour. Comparing XRD data with film morphology, the undoped film deposited at the transition temperature has a rugged surface with small (3 1 1) texture. Bisaro *et al.* [12] also observed (3 1 1) texture for the film deposited at the transition temperature which agrees with ours, except for the surface smoothness of the films. They observed the surface of partially crystalline film to be rather flat compared with that of totally crystalline film.

In order to investigate the surface morphology and grain structure, transmission electron micrographs were taken. Fig. 4 shows plane transmission electron micrographs of the films obtained at various deposition temperatures. Large grains and microcrystallites were observed at 570 °C. The HSG polysilicon is a single grain with (1 1 1) twin boundaries, as shown in the selected-area diffraction pattern (SAD). It was found that with increasing deposition temperature, the grain diameter decreases and grain density increases. From the cross-sectional transmission electron micrograph shown in Fig. 5, it was observed that small grains start to grow from the oxide substrate and a large grain protrudes from the amorphous silicon surface at 570 °C. With increasing deposition temperature, many grain boundaries impede lateral grain growth, resulting in an HSG growth at 580 °C, and further increasing the temperature results in cylindrical and/or columnar grain growth above 590 °C.

In order to examine an initial grain-growth phenomenon, the film thickness was varied. Fig. 6 shows scanning electron micrographs of 50, 100, and 200 nm thick films deposited at 580 °C. It was observed that in the 200 nm thick film, cylindrical grains observed in the 50 nm thick film disappear and an amorphous film with hemispherical grains appears. With increasing film thickness, the surface morphology changes from cylindrical to hemispherical shape and the amorphous phase increases. These results suggest that the cylindrical grain is the nucleation step which appears in an early stage of film growth.

The surface morphology was also affected by deposition pressure. Fig. 7 shows surface morphology of the films with various silane pressures at 590 °C. It was observed that with decreasing silane pressure, the shape of the grain changes from hemispherical to cylindrical. It can be said that lowering the process pressure lowers the transition temperature. It is well known that the surface silicon migration rate is enhanced when the pressure is reduced. These results suggest that the surface morphology is related to the deposition rate and silicon atom migration rate at the deposition temperature. It has been reported [13, 14]

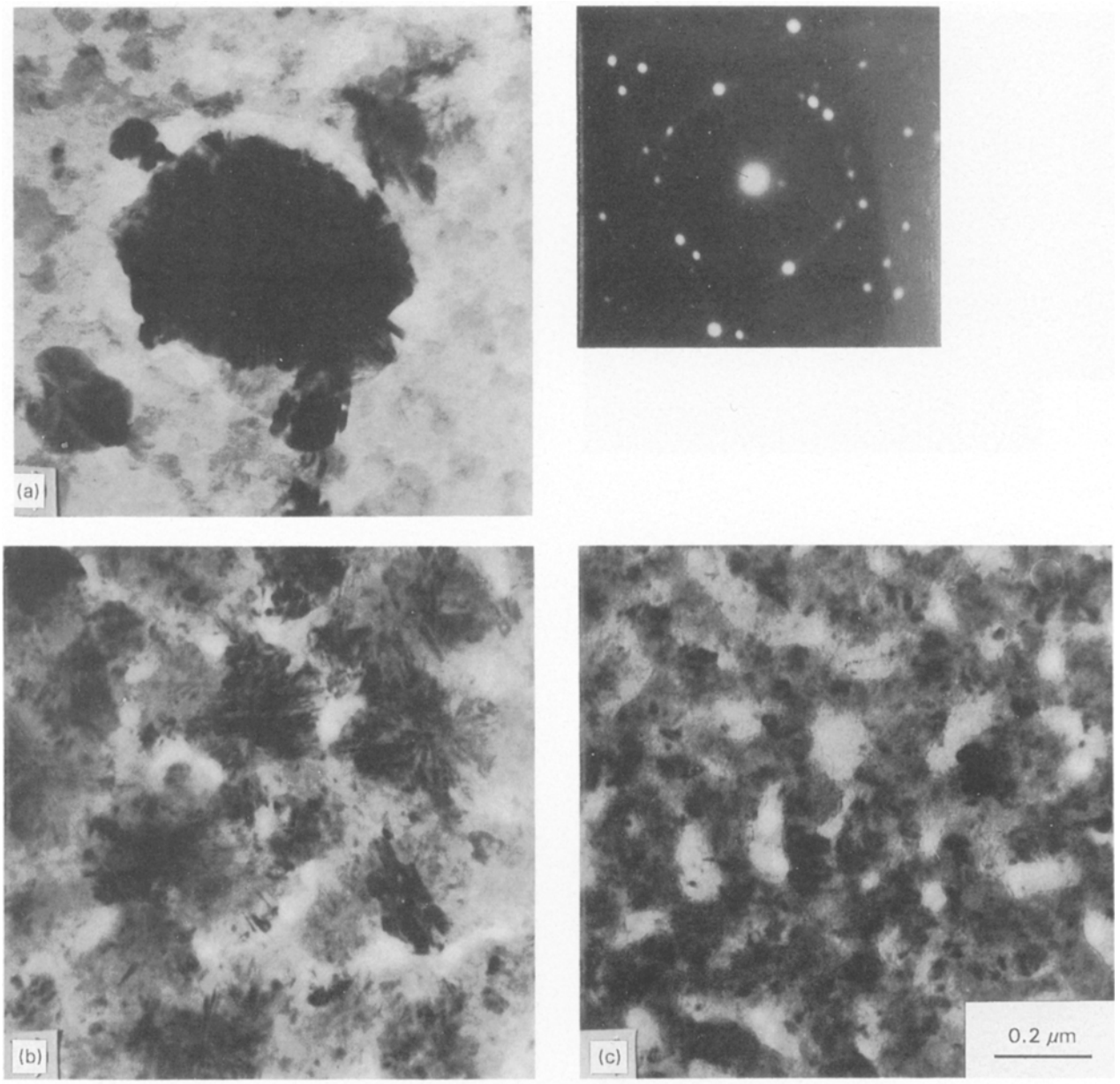


Figure 4 Plane transmission electron micrographs of films deposited at (a) 570 °C, (b) 580 °C, and (c) 590 °C. (d) The selected-area diffraction (SAD) pattern of (a).

that apparent activation energy of crystal growth rate for undoped film is about 67 kcal mol^{-1} and larger than that of the deposition rate. At a fixed temperature, a film will be partially crystallized if the crystal growth rate is comparable to the deposition rate. If the crystal growth rate far exceeds the deposition rate, the microstructure will be cylindrical and/or columnar grain structure.

Although the surface morphology of as-deposited film is almost unchanged with subsequent annealing above 1000 °C after exposuring in air, it is possible to change the morphology by *in situ* annealing without breaking the vacuum. Fig. 8 shows that cross-sectional transmission electron micrographs of the film deposited at 560 °C and annealed at 570 °C for 10 min without breaking the vacuum. In contrast to grain growth from the substrate in Fig. 5, a large grain is observed on the surface in the amorphous phase and the surface sinks in around the grain. Microcrystallites already formed during deposition are also observed on the substrate. The density of the hemispherical grains on the surface increases with increasing *in situ* annealing temperature and time. This result is in agreement with the results of Watanabe *et al.* [11] and Sakai *et al.* [15] obtained by using the ultra-high vacuum annealing of the amorphous film after removing a native oxide. They observed that the hemispherical grains are formed on the amorphous silicon surface and protrude from the original amorphous silicon plane. This result suggests that the nucleation process during annealing is different from that of deposition; grain growth starts to occur from surface silicon atom migration during *in situ* annealing, while grain growth starts from a nucleation on the substrate during deposition. It is believed that native oxide on the films exposed in air inhibits surface silicon atom migration.

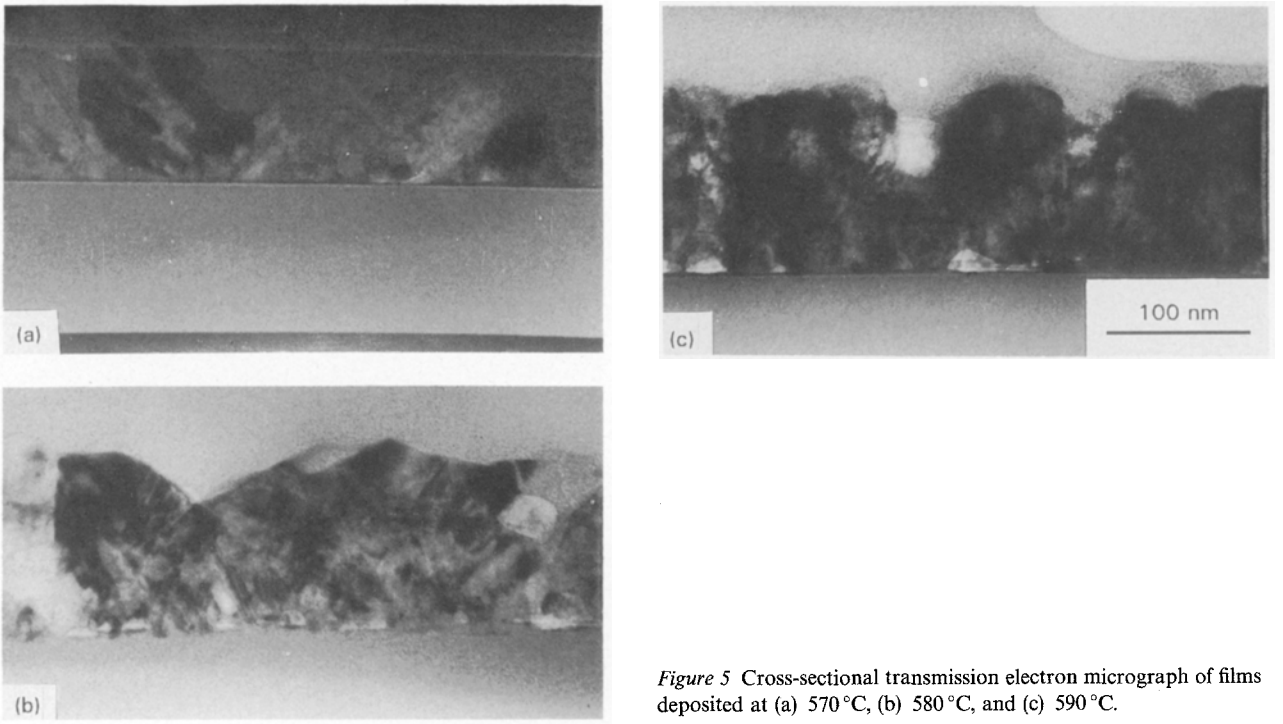


Figure 5 Cross-sectional transmission electron micrograph of films deposited at (a) 570 °C, (b) 580 °C, and (c) 590 °C.

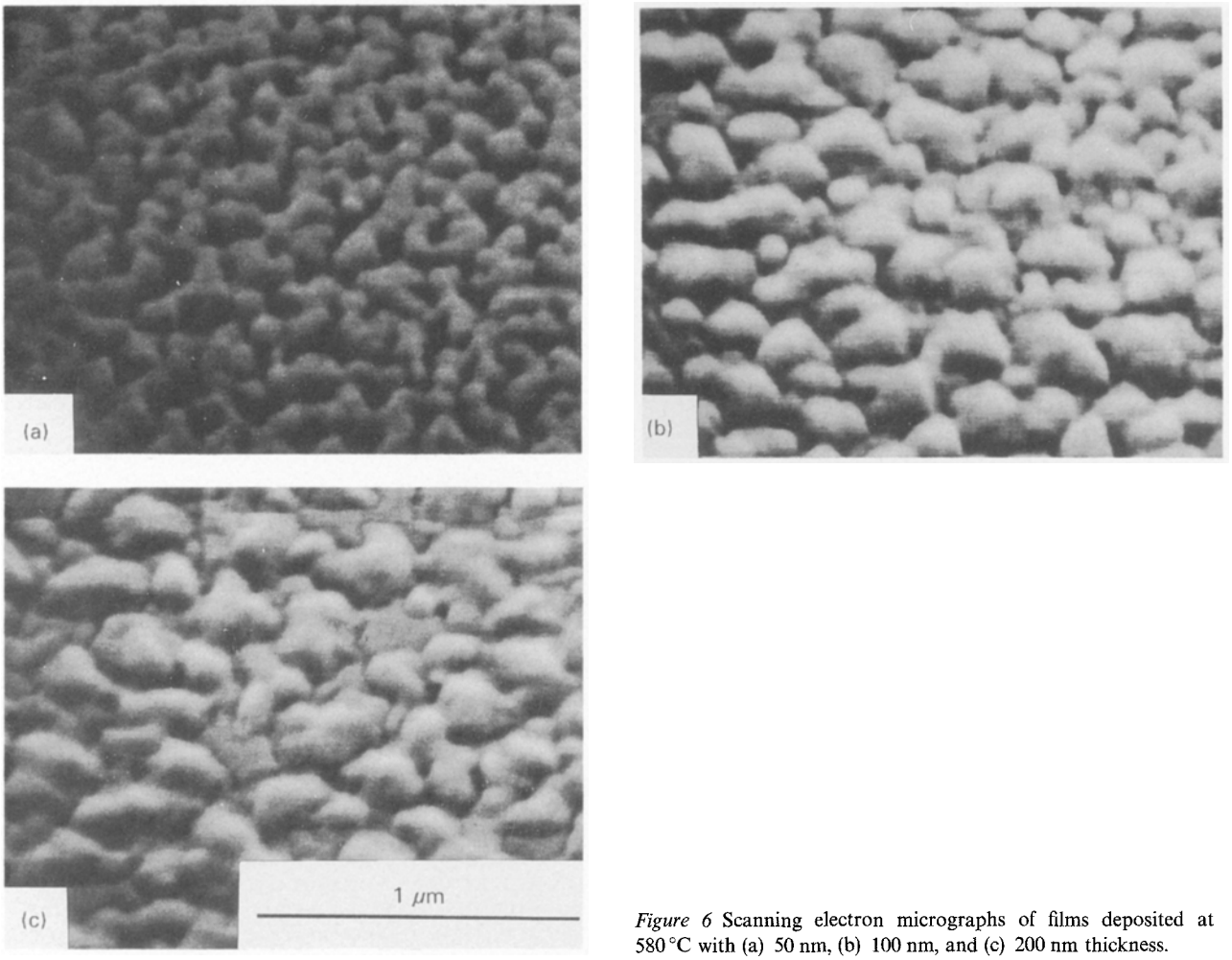


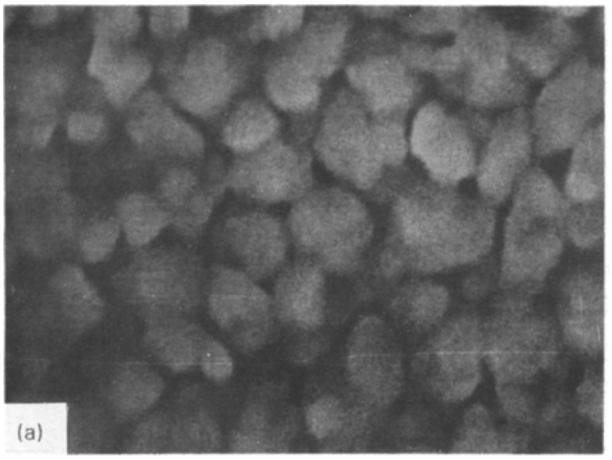
Figure 6 Scanning electron micrographs of films deposited at 580 °C with (a) 50 nm, (b) 100 nm, and (c) 200 nm thickness.

4. Conclusions

Microstructure of polysilicon films has been investigated as a function of deposition temperature, pressure, and film thickness and the results obtained are summarized as follows.

1. Film deposited at amorphous to polycrystalline transition temperature has a rugged surface with (3 1 1) texture.

2. At a fixed deposition temperature, the grain structure tends to shift from the polycrystalline to the



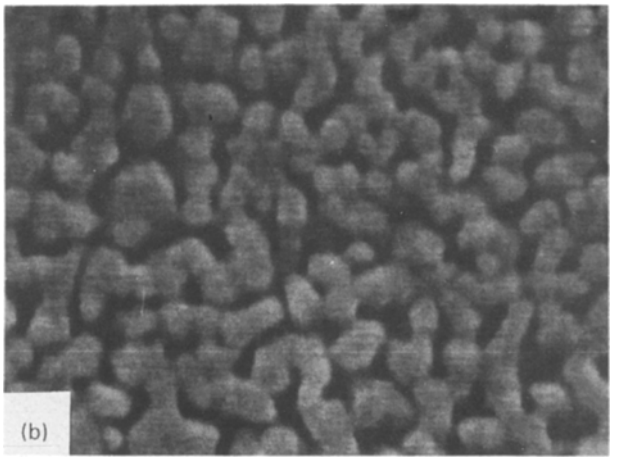
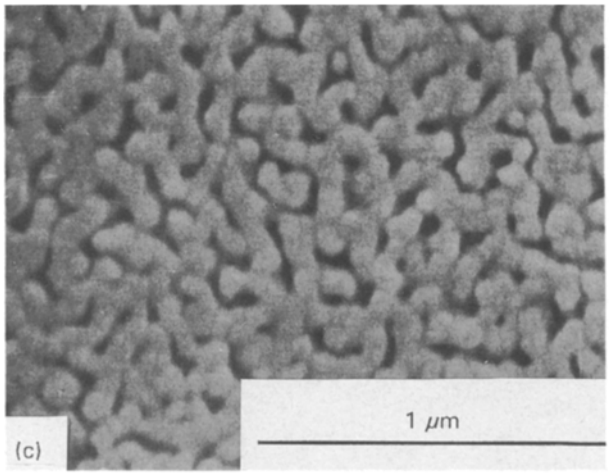


Figure 7 Scanning electron micrographs of films deposited at 590 °C under (a) 0.44 torr, (b) 0.25 torr, and (c) 0.17 torr silane partial pressure.



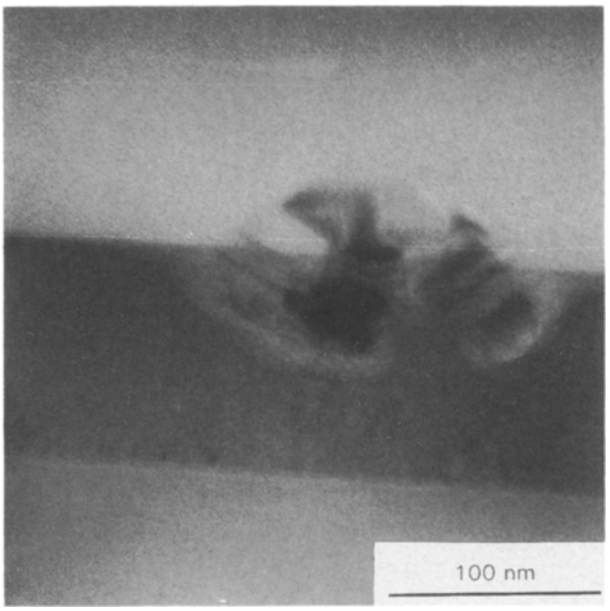


Figure 8 Cross-sectional transmission electron micrographs of films deposited at 560 °C and *in situ* annealed at 570 °C for 10 min without breaking the vacuum.

amorphous state with increasing deposition pressure and film thickness. The surface morphology and microstructure are related to deposition rate and silicon atom migration rate.

3. Nucleation during deposition starts to occur from the substrate, while nucleation during *in situ* annealing at the transition temperature without breaking the vacuum, begins to occur from surface silicon atom migration.

References

1. T. I. KAMINS, M. M. MANDURAH and K. C. SARASWAT, *J. Electrochem. Soc.* **125** (1978) 927.
2. Y. WADA and S. NISHIMATSU, *ibid.* **125** (1978) 1499.
3. T. I. KAMINS, *ibid.* **127** (1980) 686.
4. G. HARBEKE, L. KRAUSBAUER, E. F. STEIGMEIER, A. E. WIDMER, H. F. KAPPERT and G. NEUGEBAUER, *ibid.* **131** (1984) 675.
5. M. T. DUFFY and G. HARBEKE, in "Proceedings of the 9th International Conference on CVD" (1984) p. 400.
6. M. HENDRIKS and C. MAVERO, *J. Electrochem. Soc.* **138** (1991) 1466.
7. E. G. LEE and H. B. IM, *ibid.* **138** (1991) 3465.
8. T. ONO, T. MORI, T. AJIOKA and T. TAKAYASHIKI, *IEEE IEDM Tech. Dig.* (1985) 380.
9. S. MORI, Y. KANEKO, N. ARAI, Y. OHSHIMA, H. ARAKI, K. NARITA, E. SAKA GAMI and K. YOSHIKAWA, *IEEE IRPS Tech. Dig.* (1990) 132.
10. M. YOSHIMARU, J. MIYANO, N. INOUE, A. SAKAMOTO, S. YOU, H. TAMURA and M. INO, *IEEE IEDM Tech. Dig.* (1990) 659.
11. H. WATANABE, T. TATSUMI, T. NIINO, A. SAKAI, S. ADACHI, N. AOTO, K. KOYAMA and T. KIKKAWA, in "Extended Abstracts of the 23rd Conference on SSDM" (1991) p. 478.
12. R. BISARO, J. MAGARINO, N. PROUST and K. ZELLAMA, *J. Appl. Phys.* **59** (1986) 1167.
13. A. LIETOILA, A. WAKITA, T. W. SIGMON and J. F. GIBBONS, *ibid.* **53** (1982) 4399.
14. R. BISARO, J. MAGARINO, K. ZELLAMA, S. SQUELARD, P. GERMAIN and J. F. MORHANGE, *Phys. Rev. B* **31** (1985) 3568.
15. A. SAKAI, H. ONO, K. ISHIDA, T. NIINO and T. TATSUMI, *Jpn. Appl. Phys. Lett.* **30** (1991) L941.

Received 2 June 1992
and accepted 28 April 1993