Research Article

Nucleation and Growth Mechanism of Si Amorphous Film Deposited by PIAD

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The nanoscale Si films with the thickness of 2 nm, 5 nm, 10 nm, and 20 nm were deposited by plasma ion assisted deposition (PIAD) on glass substrate, in order to investigate the initial stage and the nucleation and growth mechanism of the Si film. The atomic force microscopy (AFM) was used to investigate the surface topography of the as-deposited Si film. The initial nucleation and growth process of the film was described. The continuous film had been already formed when the film thickness was 10 nm. The growth of the deposited Si film accorded with the Volmer-Weber growth mode.

1. Introduction

The use of surface coating for surface modification of ceramics is an important alternative as a means of extending the performance of ceramic materials in a wide range of applications [1]. The amorphous silicon (a-Si) film was deposited as surface-modification film on the SiC ceramic matrix which was applied in advanced optical systems, in order to modify the microstructure defects or pores on the substrate surface and provide an alternative surface which would make the polishing process more easily [2, 3]. Besides, the a-Si film was the precursor of the crystalline Si film which was widely used in photovoltaic industry and extensive investigations on the technique and mechanism of the crystallization were carried out [4, 5].

The a-Si has been reported to be deposited by typical physical vapor deposition (PVD), hot-wire chemical vapor deposition (HWCVD) [6], plasma-enhanced chemical vapor deposition (PECVD) [4], magnetron sputtering deposition [7], ultrahigh vacuum ion beam sputtering (UHV IBS) [8], and so on.

The PVD process which is an environmental friendly technology could prepare a compact and uniform a-Si film [9]. The PVD techniques including thermal evaporation techniques, sputtering technique, and ion assisted deposition have been used to fabricate Si modification films. In the typical process of the vacuum evaporation, the migration rate of atom on the surface of substrate is confined by the low incident energy (only the kinetic energy of thermal motion, ~1 eV), and the preferred orientation growth and shadow effect could also result in the loose structure with columnar growth and holes in the final as-deposited film [10]. To solve this problem, the thermal evaporation techniques are combined with ion sources which provide additional energy and momentum to influence the properties of the as-deposited films, known as ion assisted deposition (IAD) [11–13]. To reinforce the chemical reaction between the substrate and the as-deposited materials atoms which would finally improve the compactness and the adhesive strength of the deposited film, the plasma ion assisted deposition (PIAD), as one of the ion plating method, was introduced into the deposition process. The plasma produced by the advance

plasma sources (APS) was distributed in the whole cavity, which would interact with the as-deposited material atoms to transfer high energy and to ionize the as-deposited atoms. The technological merits of this plasma source are recognized by the production of high performance coatings [14].

The microstructures of the a-Si film prepared by PIAD have been investigated and the excellent properties were proved for various applications [1, 9]. But little attention was paid to the initial stage and the growth mechanism of the a-Si film deposited by PIAD, which would be important to the microstructure and the final surface morphology of the amorphous film.

One of the reasons to this neglect is that most of the surface-sensitive analytical techniques applied in coating study could not be available for nanoscale observation. The nuclei with the size larger than 50 nm could be successfully observed by high-resolution SEM [15, 16]. The topography of the nuclei with the size of several nanometers could be clearly observed by the atomic force microscopy (AFM) [17]. So, only the atomic force microscopy (AFM) has been widely used to study the growth mechanisms and dynamics and the nanometer scale morphology of crystal surfaces, since its invention in 1986 [18].

In this work, a detailed study of the nucleation and growth mechanism of the PIAD a-Si film on silicate glass substrate has been performed. The Si films with different thickness of 2 nm, 5 nm, 10 nm, and 20 nm were prepared. The nucleation and growth process of the film was observed by AFM technique. The mean size and height of the nucleus (island) was estimated by line scan in the AFM images. The nucleation and growth phenomenon had been described and analyzed. The continuous film had been already formed when the film thickness was 10 nm, and the growth process of Si film deposited on glass substrate by PIAD was found to accord with Volmer-Weber growth mode. The deposited Si clusters with the size of 1-2 nm, as the deposition unit, were always present on the surface of as-deposited continuous Si film in the PAID process.

2. Experimental Details

The nanosilicon film was deposited on silicate glass in a box-type vacuum coating machine (Leybold APS 1104 coater, Germany) with plasma ion source assisted. The target was a high-purity polycrystalline Si source (99.999%, Special Alloy Powder Metallurgy Materials, General Research Institute for Nonferrous Metals (GRINM)). Before the deposition, the substrates were cleaned successively with water in an ultrasonic cleaner and then rinsed by acetone. All samples were performed at 200°C. The vacuum in the chamber before deposition was 2.0×10^{-3} Pa. The deposition rate remained 0.1 nm/s and the power of ion source was 6 kW during the deposition process. The mean thickness of Si film was designed to be 2 nm, 5 nm, 10 nm, and 20 nm, which was real-time monitored by a quartz crystal oscillator.

The surface topographic signatures of the substrate and the Si films were observed in detail by the atomic force microscope (AFM) (SPI3800N and 144 SPA300HV, SEIKO II, NSK Ltd., Japan) in ambient atmosphere at room temperature

in contact mode. The average height (thickness) and the size of the crystalline grain in full width at half maximum (FWHM) were estimated by the line scan in the AFM images and the surface roughness reported as peak value (PV) and root mean square roughness (RMS) was also calculated.

3. Results

3.1. The Surface Morphology of the Substrate. It is known that the properties of the substrate surface would significantly affect not only the microstructure but also the growth mechanism especially at the initial stage of the film deposition. The defect or impurity sites on the substrate surface would be the priority areas of nucleation. To investigate the growth process of the nanoscale film, the required properties of the substrate surface was even crucial.

The typical AFM images of glass substrate surface in two-dimensional (2D) and three-dimensional (3D) and the line scan profile on the image were shown in Figure 1. The surface was flat without defects or scratch. The RMS of substrate surface was 0.9 nm measured by AFM at a scan size of $400 \mu\text{m}^2$. The line scan in the image showed that the surface fluctuation was gentle and the PV was about 1 nm, which means the deposition films which were designed with more than 2 nm thick in this work would be obviously observed on this substrate.

3.2. The Surface Morphology of the Si Film. Si films with the thickness of 2 nm, 5 nm, and 10 nm were deposited on glass substrate and the AFM topographic images of the surfaces were shown in Figure 2. Figures 2(a), 2(c), and 2(e) were the 2D surface topography of the 2 nm, 5 nm, and 10 nm Si film, respectively, and Figures 2(b), 2(d), and 2(f) were the 3D images of the selected area in the corresponding 2D images.

The light spheroids randomly dispersed on the surface of the 2 nm Si film in Figure 2(a) which could associate with the spheroids observed in Figure 2(b). This surface topography signature which was quite different from the surface of substrate could be induced by the deposition process. And the spheroids/protuberances could be the nucleus of Si crystallization at the initial stage of the film forming. In Figures 2(c) and 2(d) which showed the topography of the 5 nm Si film, the density and the size of the dispersed nanocrystallites, which have been confirmed in the 2 nm film, were obviously increased. As the thickness of the Si film increased to 10 nm, the surface topography became smooth without nanocrystallites structure which has been seen in the 2 nm or 5 nm film; besides more and smaller spheroids were dispersed in Figure 2(f).

3.3. The Line Scan Profile of the Nuclei. The line scan profiles of the Si film with different thickness were shown in Figure 3, and the mean height and full width at half maximum (FWHM) of the spheroids on the surface of the Si film with different thickness were summarized in Table 1.

The height and the FWHM of the protuberances on the 2 nm Si surface were 1.4 nm and 100 nm, respectively. When the thickness of the deposited film increased to 5 nm, the height and FWHM of the nuclei increased to 3.7 nm and

FIGURE 1: The AFM topographic image and line scan profile of glass substrate surface.

TABLE 1: The height and the FWHM of the nuclei on the surface of the Si film with different thickness.

Thickness	2 nm	5 nm	10 nm
Height	1.4	3.7 nm	0.9
FWHM	106.05 nm	164.05 nm	75.77 nm

164 nm, respectively. The Si nuclei with the intermediate state of merging were pointed out by arrows in Figure 3(c), which means that the coalescence process of the nuclei was progressively undergoing with the growth of the nuclei. The line scan profile of the film surface exhibited that the height and FWHM of the nuclei decreased to 0.9 nm and 75.8 nm instead of increasing with the film thickness, which indicated the growth stage/module of the deposited film was totally changed. The statistic results which have great dispersion indicated the grains in each early growth stage were presented on the film surface.

3.4. The Surface Roughness of Si Film. The surface roughness (PV and RMS) of the Si film with different thickness was measured at a scan size of $10\ \mu\text{m} \times 10\ \mu\text{m}$ which was shown in Table 2. The surface roughness increased when the Si film deposited on the substrate and with the thickness increase of the Si film from 2 nm to 5 nm, which was consistent with the results of the line scan profiles on the Si film surface. The surface roughness of the Si film with 10 nm thick was

10.2 nm (PV) and 0.8 nm (RMS) which would be comparable to the surface roughness of the substrate. The fundamental changes of the film surface roughness indicated there is no deep canyon across the whole film thickness between the independently grown-up islands. So the continues deposited film was expected.

4. Discussion

The emphases of this work were focused on the topographic signature of the Si nucleate on the substrate, the growth process, and mechanism of the nanoscale film. The whole process of the Si film growth on substrate would be described as the schematic diagram shown in Figure 4, by studying the AFM images of all the growth stages of the deposited film. The surface topography and the line scan profile of the 2 nm Si film indicated that the spheroid nuclei have been formed (Figures 2(a) and 3(a)). The nucleus shape was determined by the noninfiltration phenomenon between the substrate and the film component, which means the deposited Si atoms were more strongly bound to each other than they were bound to the substrate. This nucleation process was described as the first stage in Figure 4. The height and FWHM of the nuclei increased when the deposition thickness increased to 5 nm, which indicated the nuclei were at the growth stage. The nuclei grew up independently or by nuclei coalescence which was proved by the line scan profile in Figure 3(b) and depicted as second stage in Figure 4. The nuclei continued to grow and

FIGURE 2: The surface topography images of the Si film with different thickness. The (a), (c), and (e) were the $10\ \mu\text{m} \times 10\ \mu\text{m}$ 2D images of 2 nm, 5 nm, and 10 nm Si film respectively, and the (b), (d), and (f) were the 3D images of the selected area in the 2D images of the corresponding Si films.

FIGURE 3: The line scan profiles of the Si film with different thickness. The (a) and (b) were the profiles of the line scan marked in Figure 2(a), the (c) and (d) were the profiles of the line scan marked in Figure 2(c), and the (e) and (f) were the profiles of the line scan marked in Figure 2(e).

FIGURE 4: The schematic diagram of the deposition process of Si film on glass substrate.

coalesce until a continuous film formed which was depicted as the third stage in Figure 4. Those growth processes were accorded with the typical processes of Volmer-Weber growth mode [19, 20].

When the film thickness reached 10 nm, the surface topography was totally changed without obviously crystallize nuclei but smooth surface was observed, and the surface roughness was also reduced to be comparable with the initial substrate surface. To figure out whether the 10 nm Si film was completely continuous or not, the surface of the 20 nm Si film was also observed by AFM. The surface topography image and line scan profile were shown in Figure 5. The surface of the 20 nm film showed the same feature with the 10 nm film,

TABLE 2: The surface roughness of the substrate and deposited Si films with different thickness.

	PV (nm)	RMS (nm)
Substrate	10.1	0.8
2 nm Si film	17.3	1.0
5 nm Si film	58.1	3.0
10 nm Si film	10.2	0.8

and the profile of the grains on the surface was of 1~2 nm height which was also similar with that of the 10 nm film. There was no obvious change in the surface topography of the film with more thickness and we could conclude that the deposited 10 nm Si film was already continuous.

Besides, the nuclei with the size of 1~2 nm distributed on the continuous Si film, which were even smaller than the initial crystallize nuclei, were considered as the deposition unit in deposition process. The nanoscale cluster was refined by the impact of the plasma ions and the ionized to-be-deposited Si atoms with high energy. The nanoscale deposition units would effectively increase the interaction between the deposited Si units and realize the densification of the film which could finally improve the macroscopic properties of the deposited film.

5. Conclusion

In this work, the nanoscale Si films were deposited by plasma ion assisted deposition (PIAD) on glass substrate. Our emphases were focused on the topographic signature of the Si

FIGURE 5: The AFM topographic image and the line scan profile of the surface of the 20 nm Si deposited film.

nucleate and the growth mechanism of the film. The AFM was used to investigate the surface topography of the deposited Si film. The nucleation of the deposited Si on the substrate was observed and the spheroid nuclei were explored on the surface of the substrate at the initial stage of the film forming. As the deposition continued, the nucleus grew larger in size and the evidence of the nucleus coalescence was also shown in the line scan profile. The continuous film had been already formed when the film thickness reached 10 nm. The growth process of Si film was found to accord with the Volmer-Weber growth mode, and the deposited Si clusters with the size of 1–2 nm, as the deposition units, were always present on the surface of the as-deposited continuous Si film in the PAID process.

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