Influence of synthesis parameters and thermal annealing on grain size of polycrystalline aluminum thin film

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Abstract. The thin polycrystalline aluminium films were synthesized on monocrystalline silicon substrates by ion-plasma sputtering. The synthesis was carried out at temperatures of 80 and 160°C and deposition rate of 10 and 110 nm/min. As-deposited films were annealed for 15 h at 550°C. The morphology of aluminium films before and after annealing was obtained using SEM images. The surfaces of as-deposited Al thin films, synthesized at high temperature, were uneven, while for low temperature films they were smooth enough with Al hillocks on the top of the film. After thermal annealing, morphology of the films was changed slightly. XRD patterns were obtained to calculate the average Al grain size of as-deposited and annealed films. The XRD analysis showed that an increase in the synthesis temperature leads to an increase in the average grain size from 50 to 84 nm and that increase in the rate of Al film synthesis leads to an increase in the average grain size from 50 to 63 nm. As the result of annealing, the average grain size increased for all samples and the final meaning was from 78 to 140 nm.

1. Introduction
Polycrystalline silicon (poly-Si) thin films on foreign inexpensive substrates attract interest for using in microelectronics and solar cell applications [1]. The most widely used methods of poly-Si thin film preparation are solid-phase [2], laser-induced [3] and metal-induced [4] crystallization of thin films of amorphous silicon (a-Si). Metal-induced crystallization (MIC) of a-Si thin films provides relatively low temperatures and annealing times because of using metals (aluminum, gold, silver) as a crystallization process catalyst. The most widespread metal for realization of MIC process is aluminum (Al).

Aluminum induced crystallization (AIC) of a-Si thin films allows production of thin films of poly-Si with a large grain size at low annealing times and temperatures below the eutectics temperature of “Al/Si” system of 577°C. Nowadays, increasing the average Si grain size is an actual technological goal, since it is believed that the small grain size is responsible for the low performance of solar cells [5]. This is because recombination of free charge carriers occurs at the boundaries of poly-Si grains, and an increase in the average Si grain size leads to a decrease in the grain boundaries area, and, consequently, improves efficiency of elements of this type. It becomes possible to achieve this with an increase in the average grain size of polycrystalline Al (poly-Al), since the authors of many works [6] have shown that the formation of a poly-Si grain begins at the boundaries of Al grains.

The most technologically simple method to increase the average grain size of poly-Al thin film is thermal annealing at temperatures close to the melting point of Al (660°C). In addition, one of the
ways to increase the grain size of poly-Al is to vary such parameters of the synthesis of the Al film as deposition rate and temperature.

Moreover, Al is one of the most widely used materials for microelectronics due to its low resistivity and good adhesion to silicon and its oxide. In addition to its outstanding electrical conductivity, Al is characterized by low cost and high stability; as a result, thin films of Al and its alloys are widely used in electronic devices. Due to the high reflection coefficient and good adhesion to glass, Al is one of the most commonly used coatings for the production of optical mirrors [7]. Among of other applications of thin Al films there are near-field fiber-optic probes [8], thin-film transistors [9], flat panel displays [10], and solar elements [11].

It is known, that the structural, electrical and optical properties of thin Al films are affected by several synthesis parameters, namely the type of substrate [12], film thickness [13], deposition rate [14], and synthesis temperature [15], as well as the conditions of subsequent annealing [16]. The deposition rate is one of the most significant parameters, which could affect the structure and surface morphology of the thin films. In [17], the authors studied Al thin films on quartz substrates, which were obtained by evaporation from a boat at a rate from 6 to 120 nm/min, and it was shown that the film obtained at a high deposition rate has a larger grain size, but less roughness than the film sputtered at a low deposition rate. However, in [18] the authors showed that, with an increase in the deposition rate from 30 to 120 nm/min, the surface roughness of Al films with a thickness of 200 nm was not changed. It is shown in [19] that the mean grain size of thin Al films increased from 20 nm to 70 nm with an increase in the deposition rate.

The influence of substrate temperature on the microstructural and topographical properties of Al thin films deposited by RF magnetron sputtering at various substrate temperatures (from 40 to 100°C) is investigated in [20]. It is shown that at 80°C the diffraction is determined by (200), while at 100°C the crystal plane (111) is the dominant.

The annealing process in [21] led to changes in the surface morphological of the films, increasing the grain size and decreasing the roughness. And when the films annealed from 100 to 400°C, the resistivity of the films depended on roughness and when annealed from 400 to 500°C, the resistivity showed dependence on grain size.

In this work, we investigate the structure and surface morphology of Al thin films produced by ion-plasma sputtering on monocrystalline silicon substrates as a function of deposition rate and temperature, before and after thermal annealing.

2. Experimental details

The thin Al films were synthesized on monocrystalline silicon substrates by ion-plasma sputtering with a DC power supply at a power of 400 W. All the coating processes were carried out in a chamber in vacuum at $10^{-3}$ Pa using argon gas (99.99%). The substrates with a size of 12x18 mm were used for deposition. The synthesis was carried out at temperatures (T) of 80 and 160°C. The maximum deposition rate (V) was 110 nm/min, the minimum rate was 10 nm/min. A quartz microbalance was used to control the film thickness (about 250 nm) and deposition rate. After deposition of Al thin films, the samples were subjected to annealing for 15 h at temperatures of 550°C in a high-vacuum furnace Nabertherm RHTC 80-710/15 with a residual gas pressure of $10^{-4}$ Pa.

The SEM imaging of the deposited Al thin films was performed by a JEOL JSM-6700F scanning electron microscope. The X-ray diffractometer (Shimadzu XRD-7000) with CuKa radiation was used for phase identification and grain size quantitative measurements. The line Al (111) was chosen for determining average Al grain sizes according to full width at half maximum of the corresponding X-ray diffraction line using the Scherrer equation [22].
3. Results and discussion
SEM top view images of as-deposited Al thin films on c-Si with different synthesis parameters are shown in figure 1. For films, deposited at low temperature (Figure 1a, b), the surface is fairly uniform. It is worth noting that the separate pieces (hillocks), represented by light spots, are observed on the film surface. With an increase of synthesis temperature to 160°C for a high deposition rate (Figure 1d) the surface becomes uneven, what is confirmed further by SEM cleaved cross-sections of as-deposited Al thin films (Figure 2d).

![SEM images](image1.png)

**Figure 1.** SEM top views of as-deposited Al thin films on c-Si (100) with different synthesis parameters: (a) T = 80 °C and V = 10 nm/min; (b) T = 80 °C and V = 110 nm/min; (c) T = 160 °C and V = 10 nm/min; (d) T = 160 °C and V = 110 nm/min.

The SEM images in figure 2 show various layer thicknesses and surface morphology of as-deposited Al thin films on c-Si. The average thickness of the films was 252 nm. It can be seen that at low deposition rate and synthesis temperature the Al film is dense and uniform (Figure 2a). A similar morphology can be observed for the film (Figure 2b, c) with the synthesis parameters T = 80°C, V = 110 nm/min and T = 160°C, V = 10 nm/min.

![SEM images](image2.png)
Figure 2. SEM cross-sections of as-deposited Al thin films on c-Si (100) with different synthesis parameters: (a) T = 80 °C and V = 10 nm/min; (b) T = 80 °C and V = 110 nm/min; (c) T = 160 °C and V = 10 nm/min; (d) T = 160 °C and V = 110 nm/min.

Figure 3 shows the XRD patterns of the as-deposited samples. The films are polycrystalline and the peaks at 38.55° and 44.83° correspond to the Al (111) and Al (200) planes, respectively, according to the International Centre for Diffraction Data (ICDD) No. 00-004-0787. Peaks corresponding to Al₂O₃ (ICDD No. 99-000-3962) are not detected, indicating that any oxide layers are negligible.

The grain size obtained for these films is from 50 to 84 nm, which is consistent with the Al crystallite size obtained in [23], where the Al film is deposited on steel using a sputtering method and the grain size reaches up to 40 nm. It is worth noting that the average Al crystallite size increases from 50 to 63 nm with the growth of the deposition rate from 10 to 110 nm a synthesis temperature of 80°C. In addition, an increase in the synthesis temperature from 80 to 160°C also results in a growth of grain size from 50 to 60 nm for the deposition rate of 10 nm/min and from 63 to 84 nm for a deposition rate of 110 nm/min.

Figure 3. XRD patterns for the as-deposited polycrystalline Al thin films.

In the result of annealing, the film surface changed slightly, as it is shown in the SEM top view images of the poly-Al thin films (Figure 4). It is possible to observe hillocks for a low synthesis temperature and non-uniform morphology for a high one.
Figure 4. SEM top views of the thin poly-Al films on c-Si (100) after annealing, with different synthesis parameters: (a) $T = 80 ^\circ C$ and $V = 10$ nm/min; (b) $T = 80 ^\circ C$ and $V = 110$ nm/min; (c) $T = 160 ^\circ C$ and $V = 10$ nm/min; (d) $T = 160 ^\circ C$ and $V = 110$ nm/min.

Figure 5 shows the XRD patterns of the annealed samples and the peaks corresponding to Al (111) and Al (200) in the pattern are preserved. Intensity of the peaks corresponding to Al in the pattern does not change. Moreover, the pattern shows characteristic peak at 44.9º, which probably corresponds to the (202) planes of the a-Al$_2$O$_3$ phase (JCPDS No.29–0063) [24]. After annealing, the Al grain size increased for all samples and the final value was from 78 to 140 nm.

Figure 5. X-ray diffraction patterns for the poly-Al thin films after annealing.
4. Conclusion
The SEM results showed different morphology of thin films obtained at different synthesis rates and temperatures. Al films with dense uniform structure were obtained at a synthesis temperature of 80°C and a deposition rate of 10 nm/min. With an increase in a synthesis temperature to 160°C and deposition rate to 110 nm/min, the Al film consisted of hillocks, which grew from the substrate surface. Annealing at a temperature of 550°C for 15 hours did not lead to significant changes in the morphology of the films. The XRD method was used to obtain values for the size of the Al crystallite from 50 to 84 nm for deposited films and from 78 to 140 nm for films after annealing. An increase in the temperature or the rate of synthesis of Al films led to an increase in the size of the crystallite.

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