LARGE-GRAINED POLYCRYSTALLINE SILICON THIN-FILM SOLAR CELLS USING AIC SEED LAYERS

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ABSTRACT

Large-grained polycrystalline silicon (poly-Si) films were prepared on foreign substrates by epitaxial thickening of seed layers. The seed layers were formed by the aluminum-induced layer exchange (ALILE) process which is based on aluminum-induced crystallization (AIC) of amorphous silicon (a-Si). The epitaxial thickening was carried out at two different temperature regimes (low- and high-temperature approach). Using these large-grained poly-Si films first thin-film solar cells have been prepared. The best poly-Si thin-film solar cell obtained so far has reached an efficiency of 4.5% (high-temperature approach).

INTRODUCTION

The fast growing photovoltaic market is dominated by wafer-based crystalline silicon solar cells. It is expected that on the long term thin-film solar cells on foreign substrates will dominate the market due to their higher potential for cost reduction. Especially thin-film solar cells based on Si are attractive because of the non-toxicity and availability of the material. To overcome the limits of the currently available Si thin-film solar cells based on amorphous and/or microcrystalline Si the material quality has to be improved. Large-grained polycrystalline Si (poly-Si) characterized by a grain size much larger than the film thickness seems to be a suitable material for high efficiency Si-based thin-film solar cells on foreign substrates. Several concepts have been investigated to prepare such poly-Si films. In this paper we report on a two step approach which is called 'seed layer concept': Firstly, a very thin large-grained poly-Si film (seed layer) is prepared on a foreign substrate. Secondly, this seed layer is epitaxially thickened. The poly-Si film grown on the seed layer can be used as absorber of a solar cell. The structure of the poly-Si thin-film solar cells investigated here is schematically shown in Fig. 1. It consists of a p^+-type seed layer (which acts as a back surface field, BSF), a p-type absorber and an n^+-type emitter. Sometimes an additional BSF layer (p^+) is grown between seed layer and absorber. Regarding the preparation processes of the solar cells we follow two different concepts: (1) a low-temperature approach where all processes are carried out on inexpensive glass substrates and (2) a high-temperature approach using ceramic substrates (e.g. alumina). Due to the fact that epitaxial growth of Si at high temperatures is much easier than epitaxy at low temperatures, it is expected that the high-temperature approach leads to better results on the short term. But on the other hand the low-temperature approach features a higher potential for long term cost reduction. In this paper we describe the main results and the present state of the development of large-grained poly-Si thin-film solar cells using these two approaches.

SEED LAYERS

The p^+-type seed layers were prepared on both types of substrates (glass and ceramic) by the aluminum-induced layer exchange (ALILE) process which is based on aluminum-induced crystallization (AIC) of amorphous silicon (a-Si) [1]. The starting point for the ALILE process is a substrate/Al/a-Si layer stack containing a very thin oxide layer between Al and a-Si layer. The oxide layer is usually formed by exposure of the Al-coated substrate to air prior to the a-Si layer deposition. It plays a significant role acting as a permeable membrane during the ALILE process [2]. Annealing of the initial layer stack at temperatures below the eutectic temperature of the Al/Si system (T_{eu} = 577°C) leads to a layer exchange and a concurrent crystallization of Si resulting in a substrate/poly-Si/Al(+Si) layer stack. The thickness of the poly-Si film, which we call AIC seed layer, is determined by the thickness of the initial Al layer. The higher the

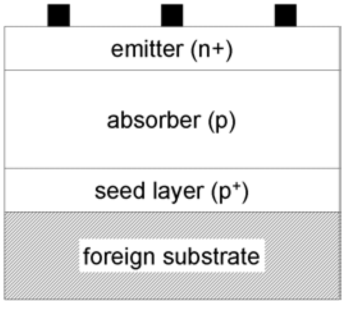

Fig. 1: Schematic structure of a large-grained poly-Si thin-film solar cell on a foreign substrate.

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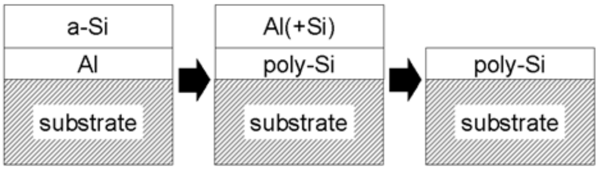

Fig. 2: Schematic illustration of the ALILE process and the subsequent removal of the Al(+Si) layer on top of the AIC (poly-Si) seed layer.

annealing temperature the faster the ALILE process. The resulting AIC seed layers feature large grains. Due to the Al incorporated in the AIC seed layer these films are always p^+-type. After the ALILE process the Al(+Si) layer on top of AIC seed layer has to be removed. The complete sequence of the AIC seed layer formation is shown schematically in Fig. 2. In the following sections specific results of the AIC seed layer formation are presented for both low- and high-temperature approach.

For the low-temperature approach, the initial layers for the ALILE process (300nm Al and 375nm a-Si) were deposited on Corning 1737F glass substrates using dc magnetron sputtering. Prior to the a-Si deposition the Al surface was oxidized for two hours to form the required permeable membrane. The annealing temperature of our standard process is 500°C. The grain size of the resulting AIC seed layers is usually up to 20µm. The crystallographic orientation of the grains was investigated by electron back-scattering diffraction (EBSD). About 60% of the area under investigation is typically tilted by less than 15° regarding the perfect (100) orientation [3]. Here we call this a preferential (100) orientation of the grains. The preferential (100) orientation is a very beneficial feature of the AIC seed layers because the quality of the Si films grown on the seed layers at low temperatures strongly depends on the orientation of the underlying grain. Due to the fact that a (100) orientation is favorable for low-temperature epitaxy, AIC seed layers are suitable even for the low-temperature approach of the seed layer concept [4]. A drawback of the ALILE process is that an Al(+Si) layer containing ‘Si islands’ (which originate from the excess of Si in the process) remains on top of the AIC seed layer. This layer has to be removed completely to obtain an optimum AIC seed layer surface for subsequent epitaxy. Especially for the low-temperature approach a complete removal of this layer is essential. Several techniques have been investigated. The best results so far were obtained by chemical mechanical polishing (CMP) using a slurry based on silicic acid. Fig. 3 shows a scanning electron microscopy (SEM) image of the surface of an AIC seed layer after removal of the Al by a wet-chemical etching process (main image). The remaining ‘Si islands’ on top of the AIC seed layer are clearly visible. Using CMP the Al(+Si) layer including the ‘Si islands’ was completely removed (Fig. 3, inset). Unfortunately CMP is probably not up-scalable at low costs for a large-area thin-film solar cell production. Therefore alternatives are investigated.

For the high-temperature approach alumina substrates (ADS996 from CoorsTek) were used. The initial layers (200nm Al and 230nm a-Si) were deposited using electron-beam evaporation. Between the two depositions the Al surface was oxidized by exposure to air for two minutes. After the deposition of the initial layer stack the samples were annealed at 500°C for a period of 30 minutes to 4 hours. Using the same conditions, the ALILE process on alumina usually leads to a smaller grain size and a higher density of ‘Si islands’ than on glass due to the much larger surface roughness of the ceramic substrates [5]. The peak-to-peak surface roughness of the alumina substrates we used is about 400nm (RMS roughness ~ 80nm). An analysis of the resulting AIC seed layers using SEM and transmission electron microscopy (TEM) revealed an average grain size between 1 and 2 µm [5]. This clearly indicates that the nucleation rate is higher when rough alumina substrates are used. To reduce the surface roughness we have covered the alumina substrates with a spin-on flowable oxide (FOx-23 from Dow Corning). This effectively lowers the peak-to-peak surface roughness to around 100nm. As a result the density of ‘Si islands’ decreases drastically while the average grain size increases to about 5 µm [5], even though there are still a lot of very small grains. Although the substrate surface is still not completely smooth, the use of the spin-on flowable oxide leads to continuous AIC seed layers that resemble those on glass except for the smaller grain sizes.

EPITAXIAL THICKENING

At low temperatures, Si films were epitaxially grown on AIC seed layers on glass with a growth rate of up to 1.2µm/h by electron-cyclotron resonance chemical vapor deposition (ECRCVD) using silane diluted in H_2. In order to grow p-type layers diborane was added to the gas flow. All depositions took place at about 580°C. As mentioned above the crystallographic orientation of the underlying

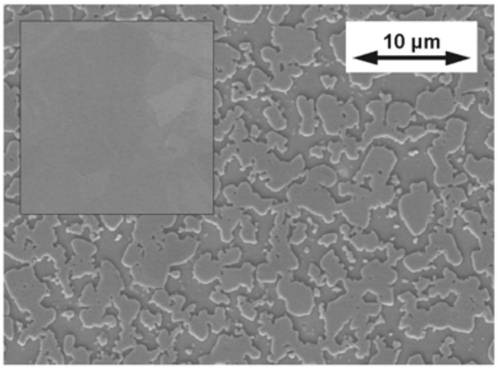

Fig. 3: Scanning electron microscopy image of the surface of an AIC seed layer after removal of Al by a wet-chemical etching process (main image) and after removal of the complete Al(+Si) layer using a chemical mechanical polishing process (inset). The scaling shown is valid for both main and inset image.

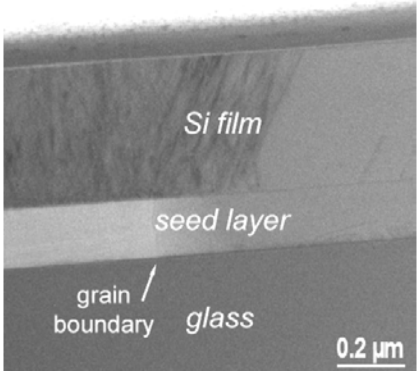

Fig. 4: Transmission electron microscopy cross section image of a Si film grown by ECRCVD on an AIC seed layer on glass.

grain has a strong influence on the quality of the grown Si film. This is clearly visible in Fig. 4 which shows a TEM cross section image of a non-intentionally doped Si film grown by ECRCVD on a glass/AIC seed layer sample. Two AIC seed layer grains are shown (the corresponding grain boundary is marked). The surface orientation of these grains was determined by analysis of the electron diffraction patterns. The surface of the grain on the right hand side is (100) orientated. This grain was epitaxially thickened. The surface of the grain on the left hand side is tilted by 7.8° with respect to (100). On this grain finecrystalline Si growth took place. This shows clearly the challenges of low-temperature epitaxy and the importance of the crystallographic orientation of the seed layer grains.

At high temperatures, the epitaxial thickening of the AIC seed layers took place by atmospheric pressure CVD using trichlorosilane diluted in H_2. The depositions were carried out at a temperature of 1130°C with a growth rate of 1.4µm/min. In-situ doping with B was done by adding diborane to the gas flow. In this way, double layers of p^+-type Si (BSF) and p-type Si (absorber) with different thickness ratios were grown on the p^+-type AIC seed layers. The additional BSF layer was deposited to reduce the series resistance of the final thin-film solar cell. The 'Si islands' were not removed from the AIC seed layers prior to the epitaxial deposition (only the Al was removed). The epitaxial quality is very good since the epitaxial layer cannot be distinguished from the AIC seed layer in cross section TEM images. The crystallographic orientations are completely reproduced [5]. Furthermore high-resolution TEM (HRTEM) investigations show a very low defect density. To investigate the grain size distribution of the epitaxial layers, the samples were mechanically polished followed by a Secco defect etch. This enabled us to visualize the grain boundaries of the layers using an optical microscope. Figure 5 compares optical images after polishing and Secco etch of the surface of absorber layers grown on (a) an alumina/AIC seed layer and (b) an alumina/spin-on oxide/AIC seed layer samples. The use of the intermediate spin-on oxide not only leads to larger grains in the AIC seed layers, but also in the epitaxial layers. The average grain size of the absorber layers grown on alumina/AIC seed layer samples is roughly about 2µm. The absorber layers deposited on alumina/spin-on oxide/AIC seed layer samples have an average grain size of around 5µm, with grains reaching a maximum size of up to 10µm. For both types of absorber layers, the grain size distribution corresponds well to that of the underlying AIC seed layers.

SOLAR CELLS

At low temperatures, first solar cell structures were prepared by deposition of a phosphorous-doped n^+-type hydrogenated amorphous silicon (a-Si:H) emitter using plasma enhanced chemical vapor deposition (PECVD) and an additional ZnO layer as a transparent conductive oxide (TCO) using reactive dc magnetron sputtering. The solar cell areas (4x4 mm^2) were defined by photolithography and wet-chemical mesa etching. The contacts were formed by evaporation of Al. The efficiencies of these first solar cell structures were below 1%. However, open circuit voltages of above 280 mV were observed without any further treatments (e.g. hydrogen passivation).

At high temperatures, an n^+-type emitter was formed by phosphorous diffusion from a P-doped silicon oxide. Bulk and surface of the poly-Si films were passivated in a

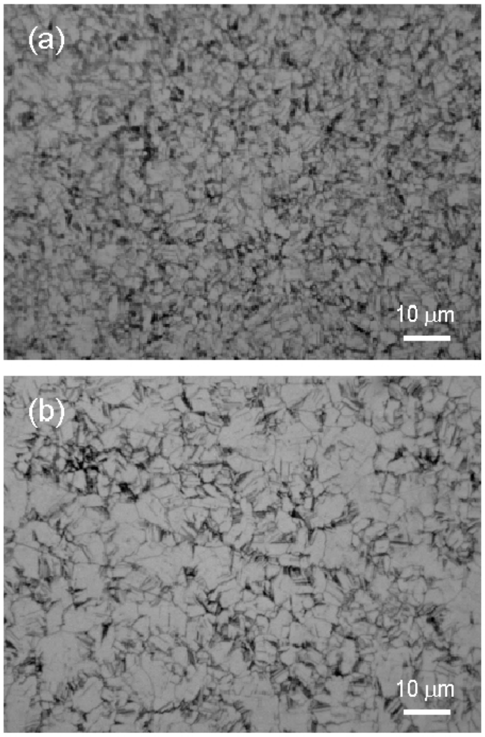

Fig. 5: Optical microscope image after mechanical polishing and Secco defect etch of an epitaxial layer grown on (a) an alumina substrate/AIC seed layer and (b) an alumina substrate/spin-on oxide/AIC seed layer sample.

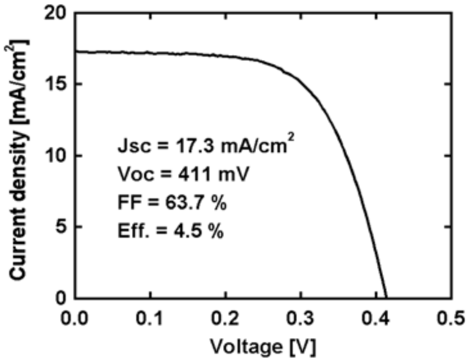

Fig. 6: Current-voltage characteristic of the best poly-Si thin-film solar cell so far (high-temperature approach).

PECVD system by plasma hydrogenation and silicon nitride deposition, respectively. By means of photolithography and wet-chemical etching mesa cells with an area of 1x1 cm^2 were formed. The absorber contact was at the periphery of the cell, while the emitter contact consists of a grid on top of the cell (this contacting scheme was also used for the low-temperature approach). A comparison between solar cells prepared on bare alumina and alumina covered with FOx showed an increase in efficiency from roughly 3.2% to 4.0% using the intermediate flowable oxide layer [6]. The highest efficiency achieved so far was on a solar cell with an absorber doping of $3x10^{16}cm^{-3}$ and a total thickness of 6µm (3µm BSF + 3µm absorber) neglecting the thickness of the AIC seed layer. The current-voltage characteristic is shown in Fig. 6. The poly-Si thin-film solar cell has an efficiency of 4.5% (J_{sc} = 17.3mA/cm^2, V_{oc} = 411mV, FF = 63.7%). By reducing the thickness of the solar cells, higher V_{oc}'s of up to 458mV were achieved [6]. Using thinner solar cells in combination with an interdigitated contact structure (reducing the high series resistance typical for the mesa cells) should therefore lead to higher efficiencies.

CONCLUSION

First large-grained poly-Si thin-film solar cells on foreign substrates were prepared by epitaxial thickening of AIC seed layers using a low- and a high temperature approach. At low temperatures, the electronic quality of the poly-Si absorber is too low. Consequently the epitaxial thickening has to be improved and the remaining defects have to be effectively passivated. At high temperatures, an efficiency of 4.5% was achieved so far. An optimization of the corresponding solar cell structure together with an increase of the AIC seed layer grain size on alumina substrates and an efficient defect passivation should result in much higher efficiencies. Considering the early state of the development the results obtained so far are mostly encouraging.

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