19th European Photovoltaic Solar Energy Conference, 7-11 June 2004, Paris, France
2AO.3.6

CRYSTALLINE SILICON THIN-FILM SOLAR CELLS ON FOREIGN SUBSTRATES: THE EUROPEAN PROJECT METEOR

S. Gall*, J. Schneider, J. Klein, M. Muske, B. Rau, E. Conrad, I. Sieber, W. Fuhs
Hahn-Meitner-Institut Berlin, Kekuléstr. 5, D-12489 Berlin, Germany
* Tel.: +49-(0)30-8062-1309, Fax: +49-(0)30-8062-1333, E-mail: gall@hmi.de

C. Ornaghi, D. Van Gestel, I. Gordon, K. Van Nieuwenhuysen, G. Beaucarne, J. Poortmans
IMEC, Kapeldreef 75, B-3001 Leuven, Belgium

M. Stöger-Pollach, J. Bernardi, P. Schattschneider
Technische Universität Wien, Wiedner Hauptstr. 8-10, A-1040 Wien, Austria

Y. Wang, O. Van Der Biest
K.U. Leuven, Kasteelpark Arenberg 44, B-3001 Leuven, Belgium

ABSTRACT: The European project METEOR aims at the development of large-grained poly-Si thin-film solar cells on foreign substrates. A two step process has been used to form the poly-Si films: (1) a thin large-grained poly-Si film (seed layer) is prepared by the aluminium-induced layer exchange (ALILE) process and (2) this seed layer is subsequently used as a template for an epitaxial thickening process. Two different concepts have been investigated: (i) a low-temperature approach using glass substrates (T < 600°C) and (ii) a high-temperature approach using ceramic substrates (T > 1000°C). The surface roughness of the ceramic substrates has a negative impact on the ALILE process. The surface roughness can be reduced by the deposition of an additional oxide layer. At high temperatures thermal CVD has successfully been used for epitaxy. At low temperatures about 73% of the area under investigation have been epitaxially thickened by a 400nm ECRCVD grown film because of the preferential (100) orientation of the seed layer. First poly-Si thin-film solar cells have been prepared at low and high temperatures. The best solar cell so far has reached an open circuit voltage of 428mV and an efficiency of 4.2%.

Keywords: Polycrystalline, Silicon, Thin-Film

1 INTRODUCTION

Thin crystalline silicon solar cells have the potential for very high efficiencies. This has been shown by the preparation of a solar cell with an efficiency of 21.5% on a thinned-down monocrystalline Si wafer with a thickness of 47µm [1]. Unfortunately this is not a real Si thin-film technology but still a Si wafer technology. It is expected that only a real Si thin-film technology, which uses large-area low-cost foreign substrates, could lead to significant cost reduction on a long term. Such a thin-film technology requires the preparation of Si layers on foreign substrates with high structural and electronic quality by an industrially applicable process. At the moment microcrystalline silicon (µc-Si:H) is the most relevant crystalline Si thin-film technology. Although reasonable efficiencies have already been reached (10%) [2] this material does not have the potential for very high efficiencies because the structural quality is relatively poor (i.e. the grain size is much smaller than the film thickness). To overcome the limits of µc-Si:H, large-grained polycrystalline silicon (poly-Si) films (i.e. the grain size is much larger than the film thickness) are necessary. Different concepts have been investigated to prepare thin large-grained poly-Si films on foreign substrates. An interesting route towards the preparation of large-grained poly-Si films at low temperatures (i.e. glass can be used as a substrate) is thermal crystallisation of amorphous silicon (a-Si:H). Module efficiencies of over 8% have been realised in a pilot production using this technology [3]. But the grain size of the Si films prepared by this technique is still in the order of the film thickness. Poly-Si films with much higher structural and electronic quality can be prepared at very high temperatures. For example an efficiency of about 16.5% has been achieved using a combination of zone-melting recrystallisation (ZMR) and subsequent epitaxial growth by chemical vapour deposition (CVD) [4]. However the ZMR process is very complex and would probably lead to excessive costs in an industrial setting.

Therefore, a low-cost concept is required which leads to high quality poly-Si films on foreign substrates. The European project METEOR (project title: 'Metal-induced Crystallisation and Epitaxial Deposition for Thin, Efficient and Low-cost Crystalline Si Solar Cells') aims to achieve this by a two step process: (1) a thin large-grained poly-Si film (seed layer) is prepared by metal-induced crystallisation (MIC) of amorphous silicon (a-Si) and (2) this seed layer is subsequently used as a template for an epitaxial thickening process.

The schematic structure of a corresponding solar cell is shown in Fig. 1. Depending on the specific process the thin large-grained poly-Si seed layer is located either on a metal-coated foreign substrate (see Fig. 1) or directly

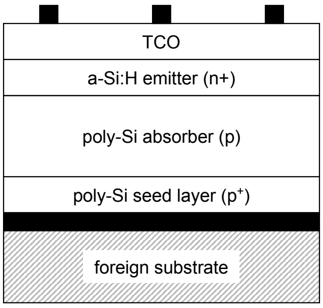

Figure 1: Schematic structure of a large-grained poly-Si thin-film solar cell on a foreign substrate.

on the foreign substrate (not shown here). The p-type absorber is grown epitaxially on the p^+-type seed layer (the seed layer consequently acts as a back surface field in this structure). Then a thin n^+-type low-temperature emitter (e.g. a-Si:H) is deposited to form the pn-junction. In case of an a-Si:H emitter a transparent conductive oxide (TCO) is necessary. Finally a metal grid is deposited onto the TCO layer. If the seed layer is located directly on the foreign substrate a metal contact to the absorber layer is needed (not shown here).

Regarding the preparation processes of the solar cells the METEOR project follows two different concepts: (i) a low-temperature approach where all processes are carried out on inexpensive glass substrates and (ii) a high-temperature approach using ceramic substrates (e.g. alumina). The low-temperature approach is characterised by a higher risk (due to the difficulties to grow epitaxially at low temperatures) but also by a larger potential for cost reduction (due to the usage of inexpensive glass substrates).

In this paper we describe the most important steps for the preparation of such a large-grained poly-Si thin-film solar cell (i.e. seed layer formation and epitaxial growth). Furthermore we present results of first solar cell structures.

2 SEED LAYER FORMATION

Thin large-grained poly-Si seed layers have been prepared using the aluminium-induced layer exchange (ALILE) process [5]. The ALILE process, which is based on the aluminium-induced crystallisation (AIC) of a-Si, starts with a substrate/Al/a-Si stack. Both layers Al and a-Si can be deposited by several techniques (e.g. sputtering). Prior to the a-Si deposition the Al-coated substrate is exposed to air to form a very thin oxide layer at the Al surface. This oxide layer plays a significant role for the ALILE process [6]. It acts as a permeable membrane controlling the diffusion of Al and Si across the interface.

Annealing at temperatures below the eutectic temperature of the Al/Si system (T_{eu} = 577°C) leads to a transformation of the initial substrate/Al/a-Si stack into a substrate/poly-Si/Al(+Si) stack. At 500°C, for example, this layer exchange usually takes much less than 2 hours.

The layer exchange begins with the local formation of Si nuclei within the initial Al layer. The growth of these Si nuclei is limited in vertical direction by both the substrate and the oxide layer at the initial Al/a-Si interface (permeable membrane). However, the growth in lateral direction is not limited by a specific interface. Therefore the Si nuclei grow until adjacent Si regions coalesce and finally form the continuous poly-Si film on the foreign substrate (the thickness of this poly-Si film - typically 200-300nm - is determined by the thickness of the initial Al layer).

The layer on top of the poly-Si film contains not only Al but also ‘Si islands’ due to the fact that the initial a-Si layer is thicker than the initial Al layer (this is necessary to prepare continuous poly-Si films). For the low-temperature approach this Al(+Si) layer is completely removed prior to the subsequent epitaxial growth, e.g. by chemical mechanical polishing (CMP). For the high-temperature approach so far only the Al is removed by wet chemical etching so that the ‘Si islands’ remain on top of the poly-Si film. The complete seed layer formation process is schematically shown in Fig. 2.

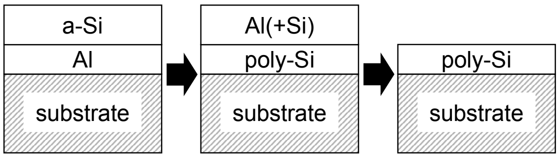

Figure 2: Schematic illustration of the ALILE process and the subsequent removal of the Al(+Si) layer on top of the poly-Si film.

We have investigated the surface of the poly-Si seed layer using electron back scattering diffraction (EBSD). The grain size of the resulting poly-Si film is up to 20µm and the surface of these grains is preferentially (100) orientated. An inverse pole figure of an EBSD orientation map of a poly-Si film on glass is shown in Fig. 3. This inverse pole figure shows clearly an agglomeration of measurement points with an orientation close to (100). About 61% of the area under investigation is tilted by less than 15° regarding the perfect (100) orientation. Here we call this a preferential (100) orientation. This feature is very important for the low-temperature approach because the (100) orientation is favourable for low-temperature epitaxy.

We have demonstrated that the ALILE process takes also place on metal-coated substrates (a continuous poly-Si film was prepared on a Mo-coated glass substrate) and is up-scalable to large areas (a continuous poly-Si film was prepared on a 3” glass substrate) [7].

Using the ALILE process we have prepared continuous poly-Si films on both smooth glass and rough alumina [8] substrates. On the alumina substrate the grain size of the seed layer is found to be much lower than on the glass substrate. This effect is related to the higher surface roughness of the alumina substrate. Therefore we have reduced the surface roughness by an additional oxide layer (FOx: flowable oxide). The FOx was deposited onto the rough alumina substrate by a spin-on process. The transmission electron microscopy (TEM) cross section image of an alumina/FOx/Al/a-Si structure (prior to the layer exchange), which is shown in Fig. 4, clearly demonstrates the smoothening effect of the FOx

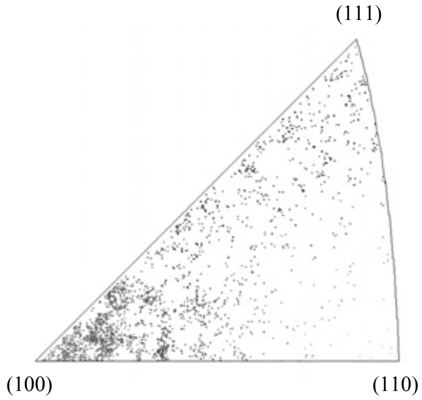

Figure 3: EBSD results (inverse pole figure) of a poly-Si film on glass showing the preferential (100) orientation of the surface.

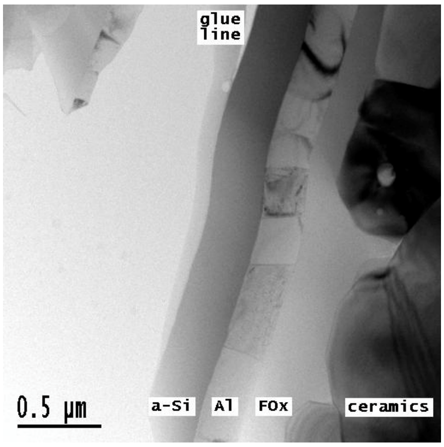

Figure 4: Transmission electron microscopy (TEM) cross section image of an alumina/FOx/Al/a-Si structure (prior to the layer exchange) showing the smoothening effect of the flowable oxide (FOx) layer.

layer. Using the FOx layer we have been able to improve the ALILE process drastically [9]. Compared to the poly-Si films on bare alumina substrates the poly-Si films on alumina/FOx substrates show much larger grain sizes (the grain sizes have increased from about 1µm to about 10µm).

The poly-Si seed layers prepared by the ALILE process are always p^+-type with an acceptor concentration of about 2 x 10^{18}cm^{-3} [10]. This is due to the fact that Al is incorporated into the poly-Si film. The p^+-poly-Si seed layer acts as a back surface field (BSF) in a $n^+/p/p^+$ solar cell structure (see Fig. 1).

We have searched for an equivalent to the ALILE process using other metals (e.g. Sb) which may result in n^+-type seed layers. This would allow the preparation of $p^+/n/n^+$ solar cells. Until now, we did not find such an equivalent process.

3 EPITAXIAL GROWTH

The large-grained poly-Si seed layers have to be epitaxially thickened to form the absorber layer of the solar cell. In contrast to the seed layer formation, where the films are prepared on different substrates but at the same annealing temperature, the growth of the absorber layer takes place in completely different temperature regimes.

3.1 Low-temperature epitaxy

For the low-temperature approach all process steps are limited to about 600°C because glass is used as a substrate. This temperature limit is very crucial to the epitaxial growth. Therefore special deposition techniques are needed which provide additional non-thermal energy to the surface of the growing film. It has been demonstrated that ion-assisted deposition (IAD) [11] and electron-cyclotron resonance CVD (ECRCVD) [12] are suitable for low-temperature Si epitaxy. It was already shown that the poly-Si seed layers prepared by the ALILE process can be epitaxially thickened using IAD [13].

Within the low-temperature part of the METEOR project we have used ECRCVD to grow the absorber layer. Prior to the ECRCVD deposition the poly-Si seed layers on glass were cleaned by a standard RCA procedure. After a final HF-dip the samples were transferred into the process chamber. During the heat-up phase the samples were held in hydrogen atmosphere. The films were grown at a substrate temperature of 585°C with a growth rate of about 20nm/min.

Fig. 5 shows an EBSD orientation map of a crystalline Si film grown by ECRCVD (about 400nm thick) on a large-grained poly-Si seed layer prepared by the ALILE process. The grey-shadowed regions correspond to areas where a crystallographic orientation of the surface could be obtained. The surface orientation of these areas is close to (100) which is known to be the favourable orientation for low-temperature Si epitaxy [12]. Within these grey-shadowed regions the underlying grains of the seed layer were epitaxially thickened. However, the black regions correspond to areas where no surface orientation could be determined (due to the finecrystalline structure of the grown Si film). The finecrystalline Si growth took place on grains of the seed layer with surface orientations which are not suitable for low-temperature Si epitaxy by ECRCVD. Analysing the EBSD orientation map shown in Fig. 5 we found that about 73% of the area under investigation was epitaxially thickened (ratio of grey-shadowed areas to the area under investigation) [14]. This is due to the preferential (100) orientation of the underlying poly-Si seed layer. To further increase the epitaxially thickened area the preferential (100) orientation of the seed layer and/or the ECRCVD deposition parameters have to be further optimised.

3.2 High-temperature epitaxy

Compared to the low-temperature process described above the epitaxial growth at high temperatures is much easier. For the high-temperature approach we have used thermal CVD at atmospheric pressure (APCVD). The Si depositions were carried out at a substrate temperature of 1130°C with a deposition rate of 1.4µm/min.

The Si growth directly on bare alumina substrates results in a grain size of the grown film below 1µm. The grain size of the grown film has been increased to about 5µm by the growth on an alumina/FOx/seed layer structure. As already mentioned above, the 'Si islands' on top of the poly-Si seed layer were not removed for the

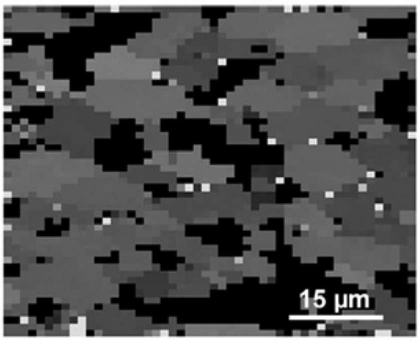

Figure 5: EBSD orientation map of a crystalline Si film grown by ECRCVD on a large-grained poly-Si seed layer prepared by the ALILE process on glass.

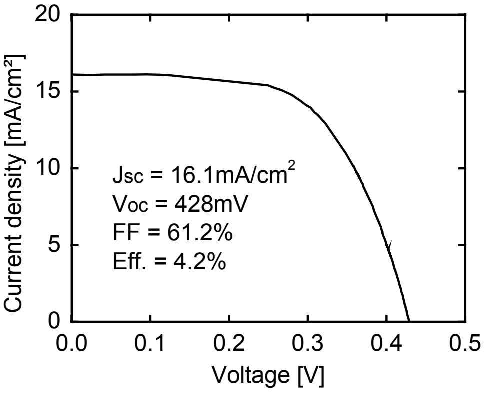

Figure 6: Current-voltage characteristic of the best poly-Si thin-film solar cell so far.

high-temperature approach. The removal of the 'Si islands' would probably lead to a further increase of the grain size.

4 FIRST SOLAR CELL STRUCTURES

First poly-Si thin-film solar cell structures have been prepared at both low and high temperatures.

The first solar cell prepared at low temperature (< 600°C) had a glass/seed layer/absorber/emitter/TCO structure. Onto the absorber layer a low-temperature emitter (a-Si:H) and a TCO (ZnO) have been deposited. The efficiency of this first solar cell structure was below 1%.

At high temperatures a much higher efficiency has been achieved using the following solar cell structure: alumina/FOx/seed layer/p^+-layer/p-layer/n^+-emitter [9]. The p^+-layer and the p-layer (each 2-3µm thick) have been deposited by thermal CVD. The n^+-emitter has subsequently been diffused from a P-doped silicon oxide. Then defects have been passivated by hydrogenation and silicon nitride deposition. After the area of the solar cell (1cm^2) has been defined by mesa etching the contacts have been evaporated (the absorber layer has been contacted only around the mesa-defined emitter area).

The current-voltage characteristic of the best solar cell so far is shown in Fig. 6. An efficiency of 4.2% has been reached (Jsc = 16.1mA/cm^2, Voc = 428mV, FF = 61.2%). The series resistance is rather high due to the above mentioned specific contact to the absorber layer. These are only first results and therefore it is expected that future improvements will lead to much higher efficiencies.

5 PERSPECTIVE

We have described first results of the European project METEOR which aims at the development of large-grained poly-Si thin-film solar cells on foreign substrates. Large-grained poly-Si seed layers prepared by the ALILE process have been epitaxially thickened at both low and high temperatures. Using this concept first solar cells with an efficiency of up to 4.2% have been prepared.

A lot of problems have to be solved to reach much higher efficiencies, especially at low temperatures because of the difficulties of the epitaxial growth.

More information on the low-temperature and the high-temperature approach within the METEOR project can be found in [15] and [9], respectively.

The METEOR project is part of an European cluster on high- and intermediate (in this paper we have called this low-temperature approach) temperature crystalline Si thin-film solar cells. Short descriptions of the involved European projects and a definition of a first common roadmap for the future development have been given in [16].

6 ACKNOWLEDGEMENTS

The work has been financially supported by the European Commission (METEOR project, contract N° ENK5-CT-2001-00543).

7 REFERENCES

[1] J. Zhao, A. Wang, S.R. Wenham, M.A. Green, Proc. 13th EPVSEC (1995) 1566.
[2] K. Yamamoto, M. Yoshimi, T. Sazuki, Y. Tawada, Y. Okamoto, A. Nakajima, Proc. 2nd WCPEC (1998) 1284.
[3] P. Basore, Proc. PV in Europe (2002) 236.
[4] A. Takami, S. Arimoto, H. Morikawa, S. Hamamoto, T. Ishihara, H. Kumabe, T. Murotani, Proc. 12th EPVSEC (1994) 59.
[5] O. Nast, T. Puzzer, L.M. Koschier, A.B. Sproul, S.R. Wenham, Appl. Phys. Lett. 73 (1998) 3214.
[6] S. Gall, J. Schneider, M. Muske, I. Sieber, O. Nast, W. Fuhs, Proc. PV in Europe (2002) 87.
[7] S. Gall, M. Muske, S. Sieber, J. Schneider, O. Nast, W. Fuhs, Proc. 29th IEEE PVSC (2002) 1202.
[8] C. Ornaghi, M. Stöger, G. Beaucarne, J. Poortmans, P. Schattschneider, IEE Proc. Circuits, Devices and Systems 150 (2003) 287.
[9] G. Beaucarne, D. Van Gestel, I. Gordan, K. Van Nieuwenhuysen, C. Ornaghi, J. Poortmans, M. Stöger-Pollach, P. Schattschneider, Proc. 19th EPVSEC (2004).
[10] O. Nast, S. Brehme, S. Pritchard, A.G. Aberle, S.R. Wenham, Solar Energy Materials & Solar Cells 65 (2001) 385.
[11] R.B. Bergmann, C. Zaczek, N. Jensen, S. Oelting, J.H. Werner, Appl. Phys. Lett. 72 (1998) 2996.
[12] J. Platen, B. Selle, I. Sieber, S. Brehme, U. Zeimer, W. Fuhs, Thin Solid Films 381 (2001) 22.
[13] N.P. Harder, T. Puzzer, P.I. Widenborg, S. Oelting, A.G. Aberle, Crystal Growth & Design 3 (2003) 767.
[14] B. Rau, I. Sieber, J. Schneider, M. Muske, M. Stöger-Pollach, P. Schattschneider, S. Gall, W. Fuhs, J. of Crystal Growth (2004) accepted.
[15] W. Fuhs, S. Gall, B. Rau, J. Schneider, Solar Energy (2004) accepted.
[16] J. Poortmans, S. Reber, S. Gall, C. Zahedi, J. Alonso, Proc. 19th EPVSEC (2004).