A ZONE MELTING RECRYSTALLISATION (ZMR) PROCESSOR FOR 400 MM WIDE SAMPLES

A. Eyer, F. Haas, T. Kieliba [1]
Fraunhofer Institute for Solar Energy Systems ISE, Heidenhofstrasse 2, 79110 Freiburg, Germany
Phone: +49 761 4588 5261, Fax: +49 761 4588 9250, e-mail: achim.eyer@ise.fraunhofer.de

ABSTRACT: Zone Melting Recrystallisation (ZMR) is widely used to convert thin microcrystalline silicon (µc-Si) films deposited on foreign substrates by Chemical Vapour Deposition (CVD) into multicrystalline silicon (mc-Si) films with grain size similar to mc-Si ingot material. It is used in Silicon on Insulator (SOI) technology but in this paper its application to crystalline silicon thin-film solar cells is addressed. The development of a new processor called ZMR400 is reported. It is equipped with a large area heater and a zone heater that uses focussed lamp radiation. The ZMR400 is powered completely by high power linear halogen lamps, thus allowing fast heating and cooling rates if required. It provides a high purity ambient (Ar, N_2, O_2) for samples up to a size of 400 x 400 x 5 mm³. Scan speeds typically range from 10 to 200 mm/min, thus leading to a maximum throughput in the range of 5 m²/h. The ZMR process can be observed and automatically controlled via CCD cameras. The quality of recrystallised silicon films depends strongly on substrate properties, on the scan speed and on the thickness of the film as demonstrated by lab scale thin film test solar cells on various substrate materials. Conversion efficiences between 9% and 13% were achieved.
The processor can also be used for other film materials, for zone melting processes of sheet-like samples, and for homogeneous large area heating processes up to temperatures of about 1500°C.
Keywords: Zone Melting, Recrystallisation, Si-films

1 INTRODUCTION

ZMR allows to convert thin silicon films on foreign substrates from an amorphous or microcrystalline grain structure into a coarse-grained multicrystalline structure. It is widely used in SOI technology but also for processing crystalline silicon thin film solar cells on various types of substrates. Of course, the processor and the process described here could also be applied to thin films of other materials. Due to its large area bottom heater the processor also allows other applications where homogeneous heating of large sheet-like samples is required, e.g. oxidation, drying, degasing, reaction bonding, sintering, laminating etc. As the processor uses radiation of halogen lamps as heat sources very fast heating and cooling is possible. The maximum temperatures achievable depend on the absorption and emission properties of the materials to be processed. They are in the range of 1400–1500°C. High purity processing in various ambient gases such as Ar, N_2, O_2) at atmospheric pressure is possible.

For the application to crystralline silicon thin film solar cells we deposit at about 900°C a highly doped silicon film (1–10µm thick) by APCVD from a $SiHCl_3$ precursor onto an appropriate substrate (e.g. low cost silicon wafers such as reclaimed wafers or low cost silicon ribbons or ceramics with appropriate thermal expansion coefficient and a high temperature applicability). The substrate is usually coated by an intermediate conductive or non conductive layer (e.g. SiC, SiO_2, Si_3N_4 or combinations of these) acting as a diffusion barrier for impurities originating from the substrate. The silicon film on such foreign substrates or intermediate layers is microcrystalline, i.e. it has to be recrystallised by zone melting in order to obtain a crystal structure of sufficient quality. That means that at least for a short time the melting temperature of silicon (1420°C) is reached at the surface of the system. ZMR of silicon films has to be conducted under a capping layer of SiO_2 or a combination of SiO_2 and SiN_x. in order to avoid agglomeration (balling-up) of the melt. After removal of the capping layer the recrystallised silicon film, now called seeding film, with grain sizes in the range of several cm² is epitaxially thickened at about 1100°C by a normally doped APCVD silicon layer (about 20–30 µm thick) which acts together with the seeding layer as the base of the solar cell.

This technology has been used for thin film solar cells on different kinds of silicon and ceramic substrates since several years as reported in several papers[2–7]. It has the potential to be a cost effective technology since only 5% of silicon are used compared to standard wafer based solar cells. All these earlier experiments used a processor called ZMR100 also developed at Fraunhofer ISE. It is able to recrystallise silicon layers up to 100 x 100 mm².

In order to extend the ZMR technology to future large size solar cells we developed the new processor, called ZMR400, for 400 mm wide and 400 mm long samples which is described in detail in this paper.

2 THE ZMR400 PROCESSOR

The principle of the new processor is shown as a 3D drawing in Fig. 1. The main components are: the loading station, the reaction chamber, the bottom heater, the zone heater, and the transport mechanism. The frame, the supply stations for electricity, gas, cooling water, cooling air and the process control via a PC. The total length of the processor is 3600 mm, the width 1600 mm, and the height 2200 mm.

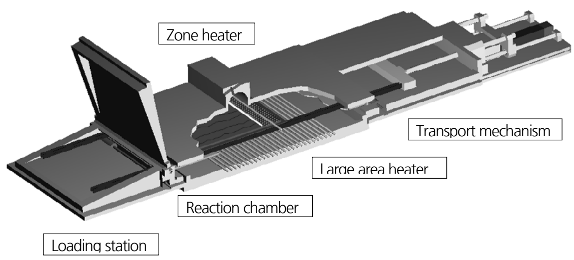

Figure 1: 3D drawing of the ZMR400 processor, total length about 2500 mm

[1] Now with ErSol Solar Energy AG, Wilhelm-Wolff-Strasse 23, 99099 Erfurt, Germany

2.1 Loading station

The processor is loaded and unloaded manually. The samples are put on a quartz plate (450 x 450 x 6 mm³) which is put on a loading mechanism and pushed into the reaction chamber manually. Fig. 2 shows a photograph of the loading station with cover opened and underneath parts of the power supply, and the gas system. A flow box above the loading station (not visible) provides high purity conditions during sample loading.

Figure 2: ZMR 400 processor, loading station and parts of the supply units.

2.2 Reaction chamber

The central part of the processor is a cold wall aluminium reaction chamber with a large quartz window (1050 x 650 mm²) at the bottom and a narrow quartz window (120 x 650 mm²) at the top. It is flanged to the loading station. The reaction chamber is gas-tight (sealed by O-rings) and can be operated under high purity conditions flooded with different gases (e.g. Ar, O_2, N_2). The inner aluminium walls are highly polished to reflect the radiation of the heating lamps. For revision the upper part of the reaction chamber can be opened pneumatically as done in Fig 3.

2.3 Large area heater

A large area heater powered by 48 linear halogen lamps is positioned underneath the bottom window of the reaction chamber. It allows a computer controlled, homogeneous, and – if needed – very fast heating-up and cooling-down of the sample up to about 1400–1500°C prior to and after the ZMR process or for other purposes. Each lamp provides 8 kW from a 650 mm long, 2.5 mm thick filament. The lamp sockets and bulbs are cooled by a strong air flow. The heated area is about 1050 mm long and 650 mm wide. It is large enough to keep the entire sample homogeneously heated during the whole process. Instead of homogeneous heating even profiles along the long axis of the lamp field are possible. The power can be set from 0–100% for each pair of lamps.

Typically a total power of about 70–80 kW is needed for a silicon film recrystallisation process.

The interior of the reaction chamber is shown in Fig. 3. Underneath the bottom window the large area heater can be seen with 28 lamps lightly illuminated for demonstration. A silicon sample of 400 x 400 mm² is lying on the quartz sample holder. The top cover of the reactor is intentionally opened here. It carries the zone heater with its single halogen lamp. The image of the large area heater is reflected from the polished inner surface of the upper cover.

2.4 Zone heater

The zone heater consists of one halogen lamp positioned exactly in the upper focus line of a 650 mm long cylindrical reflector with elliptical cross-section as shown in the principal drawing of Fig. 4. It is positioned above the narrow upper window of the reaction chamber. The lamp exhibits the same power and dimension as in the large area heater but a thinner filament (1.5 mm). The bulb and the socket of the lamp are cooled by air. Its filament can be adjusted to the focus line of the reflector using micrometer screws. The water-cooled reflector is coated with a silver foil of very high reflectivity (over 95% between 0.5–1.4 µm). Typically a power of 6–7 kW is needed for a silicon film recrystallisation process. A photograph of the zone heater is shown in Fig. 5.

Figure 3: ZMR 400 processor, opened reactor, large area bottom heater at very low lamp power for demonstration. A silicon sample of 400 x 400 mm² is lying on the sample holder. The zone heater can be seen at the top.

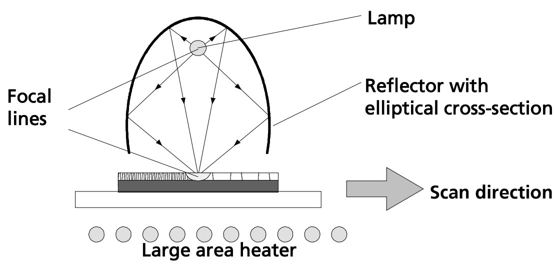

Figure 4: Principle of the zone heater above the sample and of the large area heater underneath the sample.

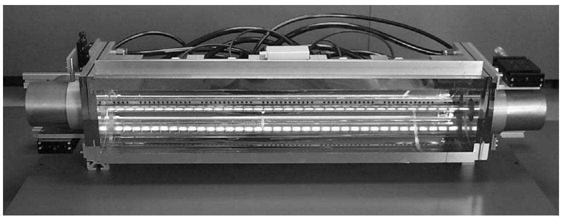

Figure 5: Zone heater from underneath; lamp at very low power for demonstration.

2.5 Sample transportation system

The sample is positioned on a quartz carrier plate (450 x 450 x 6 mm³) which is lying on two quartz rails along its edges. These rails are connected to a transportation system (motor and spindle) outside the reaction chamber (see Fig. 1) via gas-tight feed-throughs. This system moves the sample underneath the zone heater with a controlled velocity in the range of 10–200 mm/min (500 mm/min. technically possible). Thus, the molten zone is moved across the upper sample surface.

2.6 Process control

Four CCD cameras integrated into the zone heater casing allow the observation of the entire length of a 400 mm long molten zone. A fifth camera generates a microscopic image of the centre of the molten zone. It is used to detect the width of the zone by image analysis and to control the power of the zone heater lamp via a fast PID controller [3]. Thus the zone width (typically 1–1.5 mm) is kept constant within 5% during the entire ZMR process.

A close-up view of a typical molten zone is shown in Fig. 6. The upper line is the melting interface showing almost no morphology. The lower line is the facetted growth interface due to super-cooling effects. The facets lead to low angle grain boundaries in the recrystallised film.

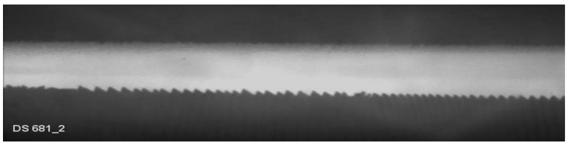

Figure 6: Close-up view of the bright molten zone during ZMR with facetted growth interface (zone width about 1 mm)

The width of the zone is extracted by digital imaging. The signal is used for an automatic closed loop control of the zone heater power in order to keep the zone width constant. The principle of the control loop is shown in Fig. 7.

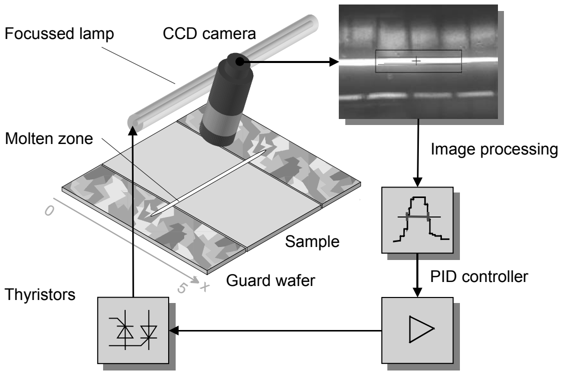

Figure 7: Closed loop control implemented in the ZMR400 processor. The width of the molten zone is measured by image analysis. The PID controller sets the power of the zone heater lamp.

3 RESULTS

3.1 Capability of the processor

The ZMR400 is powered by 49 linear halogen lamps 8 kW each but the total power is limited to 200 kW. The power supply and the cooling system is dimensioned for this power consumption over a long time period. For typical ZMR processes (10–20 µm silicon film on 0.5–1 mm thick full size silicon or ceramic substrates with a 2 µm thick SiO_2 intermediate layer) a large area heater power of 70–80 kW and a zone heater power of 6–7 kW is sufficient. This low power consumption is due to the fact that the insulating intermediate layer reduces the heat loss into the substrate quite significantly.

Zone melting of a 600 µm thick silicon wafer, i.e. moving a liquid zone of 400 x 3 x 0.6 mm³ across the wafer requires about 120 kW large area heater power and 6 kW zone heater power. Zone melting of a 2 mm thick silicon ribbon with a liquid zone of 450 x 6 x 2 mm³ required 150 kW large area heater power and 5 kW zone heater power.

3.2 Homogeneity of the molten zone

The uniformity of the zone width over its entire length (400 mm) is an important parameter for the quality and the homogeneity of the recrystallised film. It depends on several parameters like uniformity of the substrate thickness and its thermal properties as well as on the uniformity of the film thickness. The uniformity of the lamp radiation density at the sample surface is another critical parameter. It depends on the exact positioning of the lamp filament in the focus line of the reflector. It can be optimised by fine tuning the socket position via micrometer screws. Another precondition for homogeneous radiation density is the flatness (or bow) of the sample. It should be better than 2 mm over the entire zone length. A typical variation of the width of a 400 mm long zone is ±0.5 mm at a controlled mean width of 1.5 mm on perfectly flat samples.

At both ends the zone typically narrows down over a length of 10 mm due to higher heat losses along the edges of the sample. In order to compensate for these losses stripes of the same material about 20 mm wide are put along the edges of the sample. Thus the narrowing area can be reduced to about 5 mm. During the next stage of expansion and improvement of the processor additional lamps will be installed perpendicular to the existing lamps along the edges of the sample (see Section 4).

3.3 ZMR experiments

Several ZMR experiments with silicon films on different substrates were performed up to now. In order to test the influence of growth parameters like scan speed and film thickness perfect substrates were used, i.e. monocrystalline silicon wafers with polished surface coated with a 2 µm SiO_2 intermediate layer. Scan speed shows a strong impact on the final film quality as demonstrated in Fig. 7. The number of small grains and the overall dislocation density increases substantially with scan speed. On the other hand high scan speed is necessary to keep the throughput high and the process cost low. As was shown in earlier experiments the effect of scan speed also depends on the film thickness, on the solar cell process and on passivation steps. More detailed experiments are on the way. The highest efficiency realised at Fraunhofer ISE was 13.5% [6] the highest ever realised on such a system was 16.0% at Mitsubishi Electric Corp. [1].

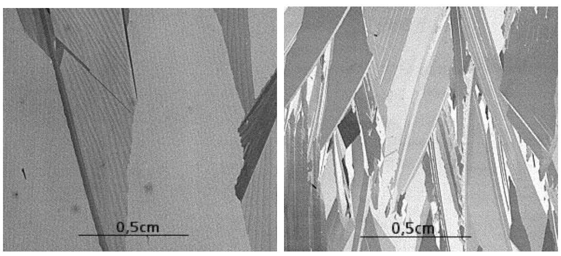

Figure 8: showing the film quality with a low scan speed of 10mm/min (left) and a high scan speed of 100mm/min (right)

Earlier experiments on different ceramic substrates like SiC, Si_3N_4, $ZrSiO_4$ and mullite demonstrated the strong influence of surface roughness and porosity compared to perfect substrates as described above. But nevertheless efficiencies between 9% and 11% were realised up to now. [2–7]

4 PLANNED EXTENSIONS OF THE PROCESSOR

Due to the encouraging performance of the new ZMR400 processor we decided to further improve the system. A second layer of linear lamps underneath the large area heater with the lamps perpendicular is already installed. It consists of 28 lamps with a lighted length of 640 mm at a total length of 1200 mm and a power of 4 kW each. This additional layer will fully compensate the temperature losses along the sample edges. Furthermore, a new sample transport system is under construction which allows in-line processing. The samples will continuously enter and leave the processor through gas curtains. We expect full operation in autumn 2004. After operating tests, the processor may be used in a future pilot plant for crystalline silicon thin film solar cells.

ACKNOWLEDGEMENT

This work has been supported by the German Federal Ministry for the Environment, Nature Conservation and Nuclear Safety (BMU) under contract No 0329850C

REFERENCES

[1] H. Morikawa, Y. Nishimoto, H. Naomoto, Y. Kawama, A. Takami, S. Arimoto, T. Ishihara and K. Namba, "16.0 % Efficiency of large area (10 cm x 10 cm) thin film polycrystalline silicon solar cell", Sol. Ener. Mater. Sol. Cells **53**, 23-28 (1998)

[2] A. Eyer, F. Haas, T. Kieliba, D. Oßwald, S. Reber, W. Zimmermann and W. Warta, "Crystalline silicon thin-film (CSiTF) solar cells on SSP and on ceramic substrates", J. Cryst. Growth **225**, 340-347 (2001)

[3] S. Reber, W. Zimmermann, and T. Kieliba, "Zone melting recrystallization of silicon films for crystalline silicon thin-film solar cells", Sol. Ener. Mater. Sol. Cells 65, 409-416 (2001)

[4] T. Kieliba, S. Bau, D. Oßwald, and A. Eyer, "Coarse-grained Si films for crystalline Si thin-film solar cells prepared by zone-melting recrystallization", EUPVSEC Munich (2001) 1604

[5] T. Kieliba, S. Bau, R. Schober, D. Oßwald, S. Reber, A. Eyer and G. Willeke, "Crystalline silicon thin-film solar cells on $ZrSiO_4$ ceramic substrates", Sol. Ener. Mater. Sol. Cells 74, 261-266 (2002)

[6] T. Kieliba, J. Pohl, A. Eyer, and Christian Schmiga, "Optimization of c-Si films formed by zone-melting recrystallization for thin-film solar cells", WCPEC3-Conference, Osaka (2003) 1170.

[7] S. Reber, A. Hurrle, A. Eyer, and G. Willeke, "Crystalline Silicon Thin-Film Solar Cells – Recent results at Fraunhofer ISE", Solar Energy 2004, in print.