

## RECENT PROGRESS IN AMORPHOUS AND MICROCRYSTALLINE SILICON BASED SOLAR CELL TECHNOLOGY

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**ABSTRACT:** This contribution discusses recent scientific and technological challenges for the development of highly efficient amorphous (a-Si:H) and microcrystalline silicon ( $\mu$ c-Si:H) based thin film solar cells. Aluminium doped ZnO films prepared by sputtering and post deposition etching serve as transparent conductive oxide (TCO) material, which provide excellent light trapping properties. Challenges are the transfer of this approach to cost-effective reactive sputtering from metallic targets and the reduction of optical absorption losses in the front TCO films. We developed  $\mu$ c-Si:H solar cells by plasma-enhanced chemical vapour deposition (PECVD) using 13.56 MHz and 40.68 MHz excitation frequency in a process regime of high deposition pressures and high RF-powers. These conditions provide sufficiently “soft” deposition for the growth of high quality  $\mu$ c-Si:H material and yield high deposition rates. A stable aperture area module efficiency of 10.1 % was obtained for an a-Si:H/ $\mu$ c-Si:H module on 10x10 cm<sup>2</sup> substrate size. In case of the  $\mu$ c-Si:H PECVD process we discuss the questions of process stability, process reproducibility and up-scaling to large area production systems.

**Keywords:** thin film, Si, ZnO

### 1 INTRODUCTION

Thin-film solar modules based on amorphous (a-Si:H) and microcrystalline ( $\mu$ c-Si:H) silicon are one of the most promising future PV technologies. These materials involve almost no ecological risk during manufacturing, operation and disposal. Low process temperatures facilitate the use of a variety of low-cost substrate materials like float-glass, metal- or even plastic-foils. Only a few micrometers of film thickness are required to absorb most of the sunlight. Several production plants with multi-megawatt capacity have already proven the manufacturability of solar modules based on a-Si:H leading to a market share of a-Si:H solar modules of 4-5 %.

The costs of production in terms of costs per Watt peak of module power are lower than for the crystalline silicon (c-Si) wafer technology, but are partly counteracted by the lower module efficiencies of 5 to 7 % as compared to 10 to 16 % in case of c-Si. In spite of the lower efficiency, a-Si:H based solar modules are successful in very different market segments offering unique advantages (e.g. for building integration [1]) and can also be produced on flexible substrates [2].

Moreover, recent progress in research and development promises to partly close the efficiency gap mentioned above. Tandem cells consisting of a-Si:H top cells and microcrystalline silicon ( $\mu$ c-Si:H) bottom cells have emerged as a new cell type and bear the potential of significantly higher module efficiencies while maintaining the advantages of the a-Si:H technology (i.e. large area processing, low process temperatures, and abundance of the raw materials). The a-Si:H/ $\mu$ c-Si:H tandem cell concept was pioneered at the University of Neuchâtel [3] using the VHF-PECVD technique. Since then, several research groups have demonstrated high efficiencies by applying different deposition techniques, light trapping concepts and substrates [3-7]. A first small scale production was started by Kaneka Co., Japan [4].

Necessary prerequisites for a cost-effective mass production of solar cells incorporating  $\mu$ c-Si:H films are the demonstration of high deposition rates and scalability to large areas. Recently, strong efforts were announced

by producers of vacuum coating equipment to develop and provide large area PECVD reactors for a-Si:H and  $\mu$ c-Si:H based solar cells [8-10]. Progress in this field will significantly reduce the production costs, and moreover, will limit the risk for possible investors. The second key challenge is the realization of a cost-effective light trapping scheme to obtain high cell efficiencies at small absorber layer thickness.

This contribution discusses the perspectives and challenges of highly efficient thin-film solar cells. We will briefly introduce our baseline PECVD process for  $\mu$ c-Si:H and a-Si:H/ $\mu$ c-Si:H solar cells on 30x30 cm<sup>2</sup> substrate size. The light trapping concept comprises surface-textured ZnO coated glass substrates and a ZnO/Ag back reflector both prepared by sputtering. We discuss the transfer of this approach to cost-effective reactive sputtering from metallic targets and show the potential for higher cell efficiencies by further reducing free carrier absorption losses in the transparent front contact. One major cost factor and technological challenge for the mass production of  $\mu$ c-Si:H based solar cells is the deposition of  $\mu$ c-Si:H i-layers on large areas at high deposition rates. We briefly discuss the questions of process stability and process reproducibility for the  $\mu$ c-Si:H baseline process, address the development of  $\mu$ c-Si:H solar cells at high deposition rates using 40.68 MHz plasma excitation frequency and, finally, discuss the up-scaling towards square meter substrate size.

### 2 EXPERIMENTAL DETAILS

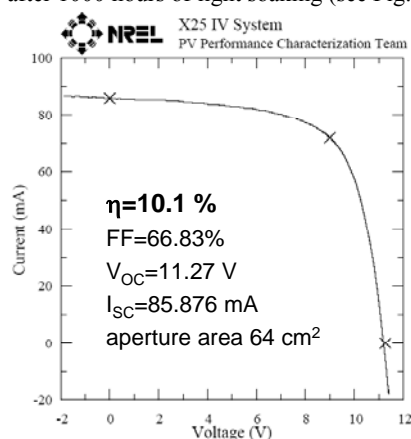
All cells and modules presented in this study were prepared in the so-called “superstrate” configuration starting from bare glass substrates. The substrate size for all films, solar cells and modules was either 10x10 cm<sup>2</sup> or 30x30 cm<sup>2</sup>. ZnO coated glass substrates prepared by magnetron sputtering and post deposition etching were applied as front TCO. These films are smooth in the as-deposited state. A textured surface is generated by wet chemical etching in diluted hydrochloric acid (HCl), which is either done manually in a chemical bath or by using an etching apparatus, especially designed for this

purpose. The ZnO films were prepared by either non-reactive RF-sputtering or reactive MF- (mid frequency) sputtering from ceramic or metallic targets, respectively, using in-line sputtering systems (see [11] for more details). All silicon layers were prepared in a PECVD reactor for 30x30 cm<sup>2</sup> substrate size [12] using either 13.56 MHz or 40.68 MHz plasma excitation frequency. A ZnO/Ag double layer serves as back contact, which was also prepared by magnetron sputtering.

The solar cell and module characterisation was performed using state-of-the-art equipment. The IV-curves under illumination (AM 1.5, 100 mW/cm<sup>2</sup>, 25 °C) were determined using a continuous light (DC) sun simulator of class A (Wacom) or a flasher (Berger). The measuring equipment allows the characterization of cells and modules with an area up to 12x12 cm<sup>2</sup> (Wacom) or 30x30 cm<sup>2</sup> (flasher) [13]. For light soaking tests the cells and modules were exposed to AM 1.5 illumination (100 mW/cm<sup>2</sup>) at open-circuit condition and a constant temperature of 50 °C. Module efficiencies refer to the aperture areas of 8x8 cm<sup>2</sup> (64 cm<sup>2</sup>) for 10x10 cm<sup>2</sup> and 26x26 cm<sup>2</sup> (676 cm<sup>2</sup>) for 30x30 cm<sup>2</sup> substrates.

### 3 BASELINE PROCESS FOR SOLAR CELL AND MODULE DEVELOPMENT

During the past years baseline processes for a-Si:H p-i-n,  $\mu$ c-Si:H p-i-n and a-Si:H/ $\mu$ c-Si:H p-i-n-p-i-n cells were established in the 30x30 cm<sup>2</sup> PECVD system yielding stable test cell efficiencies of 8 %, 8.9 % and 11.2 %, respectively. Textured-etched ZnO coated glass substrates were prepared by RF-sputtering from ceramic ZnO:Al targets. Commercially available SnO<sub>2</sub>:F from a variety of suppliers is routinely tested for comparisons and as reference. The development of device quality  $\mu$ c-Si:H films was performed using 13.56 MHz PECVD in a deposition regime of high plasma powers and deposition pressures, yielding a deposition rate of 0.5 nm/s for highest cell performance. The preparation of monolithically series connected thin-film modules was performed by high speed laser scribing for all patterning steps. So far, aperture area initial module efficiencies of 10.6 % and 10.8 % were achieved for a-Si:H/ $\mu$ c-Si:H tandem cells on substrate sizes of 30x30 cm<sup>2</sup> and 10x10 cm<sup>2</sup>, respectively (see also [14]). The efficiency of the latter small area module stabilised at an efficiency of 10.1 % after 1000 hours of light soaking (see Fig. 1).



**Figure 1:** J-V-curve of an illuminated a-Si:H/ $\mu$ c-Si:H tandem module.

Note that very recently, the a-Si:H/ $\mu$ c-Si:H baseline process was also successfully applied for the development of flexible solar modules yielding already 9 % aperture area module efficiency [15].

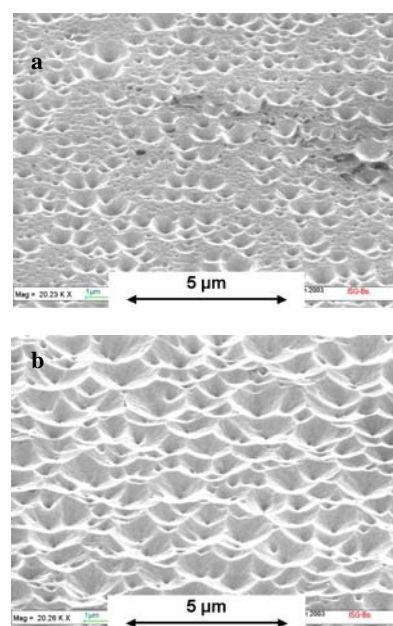
### 4 DEVELOPMENT OF SURFACE TEXTURED ZNO

In the p-i-n (superstrate) configuration the transparent conductive oxide film serves as front contact material and has to combine low series resistance, high transparency and an adequate surface texture. Examples for such TCO films are fluorine doped tin oxide films (SnO<sub>2</sub>:F) [16], boron-doped ZnO prepared by low pressure chemical vapour deposition (LPCVD) [17], intrinsic and aluminium-doped films deposited by expanding thermal plasma CVD [18], or the combination of magnetron sputtering and a post-deposition chemical etching step, which was already introduced [6,19,20].

Note, that the TCO optimization has to be done to a large extent empirically by solar cell deposition and characterization since it is very difficult to experimentally identify specific TCO properties that uniquely correlate with the measured enhancement of the short-circuit currents [21].

#### 4.1 RF-sputtering from ceramic targets

At the IPV ZnO:Al films prepared by RF-sputtering from ceramic targets serve as standard reference substrate for the  $\mu$ c-Si:H solar cell development. Fig. 2 shows how the surface morphology of such a ZnO:Al film changes upon etching in diluted hydrochloric acid. The SEM micrograph in Fig. 2a) illustrates that a short dip in hydrochloric acid leads to randomly distributed craters with diameters of less than 1  $\mu$ m, while – on a microscopic scale – parts of the ZnO surface area are only slightly attacked and remain rather smooth. After prolonged etching the crater formation proceeds until a homogeneously textured surface is obtained (see Fig. 2b).

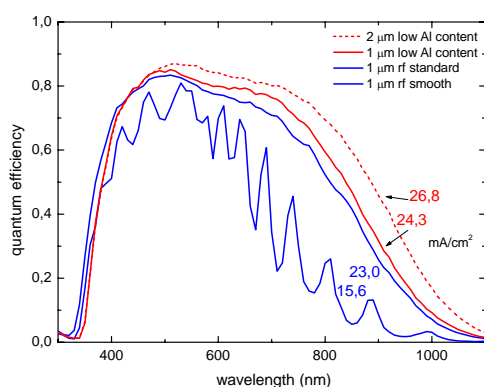


**Figure 2:** SEM micrographs of surface-textured ZnO:Al-films after short (a) and optimized etching time (b).

Due to the surface roughening, the overall short-circuit current density is about 50 % higher for the texture-etched substrate than for the smooth reference (see Fig. 3).

The potential for further current enhancement by optimized front TCO properties is nicely demonstrated by the following experiment. In case of the RF-sputtering process, we succeeded to reduce the Al-target concentration and still maintain a surface-texture, which is very similar to the example plotted in Fig. 2b). However, the sheet resistance increased, but is still below  $15\ \Omega$ . The quantum efficiency curves of two  $\mu\text{-Si:H}$  cells prepared on these optically improved substrates are shown in Fig. 3. Compared to the reference substrate, the current gain is around  $1\text{ mA/cm}^2$ . The short-circuit current densities, calculated from the spectral response measurements, are as high as  $24.3\text{ mA/cm}^2$  and  $26.8\text{ mA/cm}^2$  for an i-layer thickness of  $1.0$  and  $2.0\ \mu\text{m}$ , respectively.

As an alternative or additional feature, efficient light trapping for silicon thin-film solar cells can also be introduced by surface textured glass substrates [22,23]. In general, an optimal absorption scheme consisting of anti-reflective coatings, intermediate reflectors, front TCO films with higher carrier mobility and perfect mirrors as back reflectors can further boost the short-circuit current and thus cell efficiency (see [24] for a more detailed discussion).



**Figure 3:** Quantum efficiency curves of  $\mu\text{-Si:H}$  solar cells on different smooth and texture-etched  $\text{ZnO:Al}$  front contacts. Compared are RF-sputtered films prepared using standard and low  $\text{Al}_2\text{O}_3$  weight concentrations in the ceramic  $\text{ZnO}$  target.

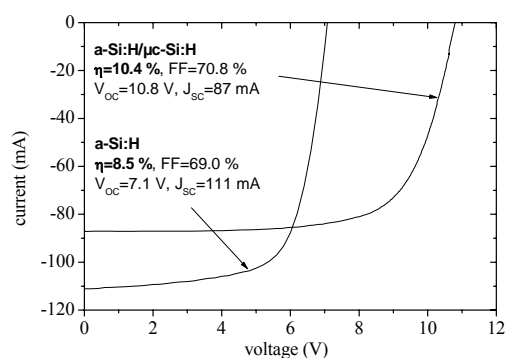
#### 4.2 High rate MF-sputtering from metallic targets

Further progress towards an application for industrial silicon thin-film solar module production requires high quality  $\text{ZnO}$  films with high uniformity on large areas to be produced at high deposition rates and low costs. This challenge of up-scaling small area  $\text{ZnO}$  films to substrate areas of the order of one square meter was taken up in a joint R&D project [25], which focuses on a high rate reactive mid-frequency (MF) sputtering process for  $\text{ZnO:Al}$  [26]. By using a MF-sputtering process described elsewhere [27] we realised dynamic deposition rates of up to  $115\text{ nm-m/min}$  (corresponding to static rates of up to  $440\text{ nm/min}$ ). Low Al doping concentrations and substrate temperatures exceeding  $200^\circ\text{C}$  had to be applied to obtain low electrical resistance and high optical transparency over a wide

spectral range. A careful optimisation and stabilisation of the working point in the transition regime between metallic and oxidic sputtering mode was further required to tune the structural film properties for the wet chemical etching process applied afterwards [27,28].

Up-scaling to  $60\times 100\text{ cm}^2$  substrate area is currently performed at the Fraunhofer Institute FhG-IST in Braunschweig. Details about the sputtering equipment and process conditions are described in related work [28]. Extra white float glass (type diamond), provided by SAINT GOBAIN GLASS served as substrate for the following experiments.

On  $10\times 10\text{ cm}^2$  and  $30\times 30\text{ cm}^2$   $\text{ZnO}$  coated glass substrates – coated in parallel at IST, we applied the baseline process for a-Si:H and a-Si:H/ $\mu\text{-Si:H}$  solar modules. So far, aperture area initial efficiencies of 8.5 % and 10.4 % were obtained for the single junction and tandem junction solar modules, respectively (see Fig. 4 for the J-V curves). On  $30\times 30\text{ cm}^2$  an initial aperture area module efficiency of 9.8 % ( $\text{FF}=71.85\%$ ,  $V_{\text{oc}}=35.5\text{ V}$ ,  $I_{\text{sc}}=260\text{ mA}$ ) was obtained. In this case the efficiency is still partly limited due to current mismatch. Optimisation is ongoing.



**Figure 4:** J-V curves of illuminated a-Si:H and a-Si:H/ $\mu\text{-Si:H}$  modules prepared on mf-sputtered  $\text{ZnO}$  coated float glass.

#### 4.3. Cost considerations

Within the TCO-project already mentioned, cost of ownership (COO) calculations for the reactive  $\text{ZnO}$  sputtering process described above, were performed by the equipment manufacturer Applied Films [29]. These calculations include the deposition of a thin  $\text{SiO}_2$  protection layer on the cleaned float glass and the preparation of the Al doped  $\text{ZnO}$  film by reactive sputtering from metallic targets. Not included are the costs for the glass substrate and for the etching process. Note that the etching process is fast and requires only very cheap chemicals in low concentration. The COO calculations are based on a large scale production machine with a coating capacity of  $400,000\text{ m}^2$  per year. Assuming a total area stable module efficiency of 8.5 % for the a-Si:H/ $\mu\text{-Si:H}$  tandem module, the calculated costs per Wp are 0.10 €. These projected costs can be further reduced considering a very large production plant and/or by increasing the solar module efficiency.

## 5 PECVD OF MICROCRYSTALLINE SILICON

The 13.56 MHz PECVD technology, with which the IPV demonstrated high efficiencies for  $\mu\text{-Si:H}$  solar cells

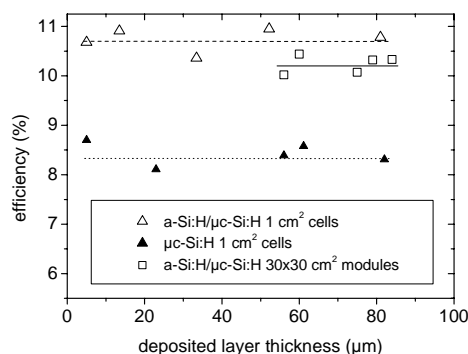
(see e.g. [6,14,30]), build the basis for the 30x30 cm<sup>2</sup> a-Si:H/ $\mu$ c-Si:H module development. In the following, we shortly summarize technological aspects of this  $\mu$ c-Si:H deposition regime. 40.68 MHz excitation frequency is studied on 30x30 cm<sup>2</sup>, because it may combine the advantages of both a high plasma excitation frequency and compatibility with the application of large area planar electrodes. Then, we present results regarding process stability of the  $\mu$ c-Si:H deposition at high deposition pressures and high plasma power densities. Finally, we briefly discuss issues related to the up-scaling of the  $\mu$ c-Si:H PECVD process to m<sup>2</sup> size.

### 5.1 Process regime at 13.56 MHz

A promising route to obtain high deposition rates for  $\mu$ c-Si:H is the use of high process pressures in combination with high discharge powers. For the 13.56 MHz regime, deposition pressures above 10 hPa were required to achieve high efficiencies of >8 % and high growth rates >0.5 nm/s [30,31].

Although our processes have shown to be reproducible and are able to yield highly efficient solar cells, there are still some issues of concern. The narrow deposition window for  $\mu$ c-Si:H is very sensitive to any drift or variation of e.g. mass flow controllers, matching properties of the high frequency power, substrate type or inter-electrode distance. Hence it follows that a careful design of the PECVD systems and an on-line control of the deposition process is mandatory.

Moreover, the high pressure and high power condition applied for  $\mu$ c-Si:H deposition lead to significant powder formation and dust is visible in the deposition chamber already after only a few runs. However, usually no powder is observed on the substrates or on the powered electrode indicating that the powder formation takes place outside the deposition zone and does not affect the  $\mu$ c-Si:H growth. To further confirm this statement, Fig. 5 shows the cell efficiency as function of the cumulative deposited layer thickness for different cell types prepared between two maintenance cycles in the 30x30 cm<sup>2</sup> PECVD system. Only cell and module efficiencies for standard 13.56 MHz  $\mu$ c-Si:H depositions are plotted which count for 20–30 % of the depositions within this operation cycle. The rest of the depositions were made at various non-standard but  $\mu$ c-Si:H conditions, can as such not be compared to one another, and are therefore not included in the graph.



**Figure 5:** Efficiencies of  $\mu$ c-Si:H based cells (cell area: 1 cm<sup>2</sup>) and a-Si:H/ $\mu$ c-Si:H modules (substrate size: 30x30 cm<sup>2</sup>) as a function of the accumulated layer thickness between two maintenance intervals.

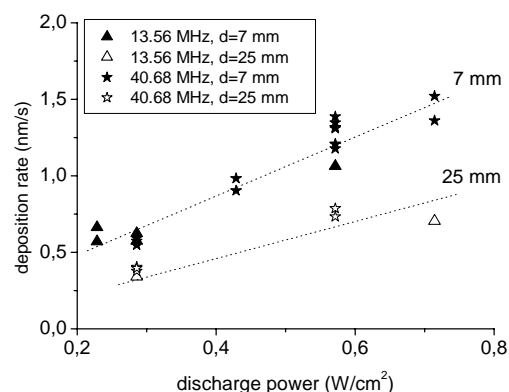
One can see that even after 80  $\mu$ m of cumulative deposited  $\mu$ c-Si:H film thickness, still highly efficient (>10 %) and shunt free 30x30 cm<sup>2</sup> modules can be prepared. Certainly this still has to be proven by long term tests in production type reactors.

### 5.2 Deposition Rate enhancement

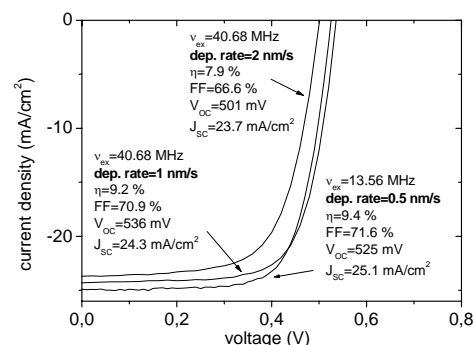
Record solar cell efficiencies at very high deposition rates have been reported for process regimes which combine plasma excitation frequencies >60 MHz and high deposition pressures [32,33]. It is reported that for discharge powers with values >1 W/cm<sup>2</sup> deposition rates >2.0 nm/s can be achieved [34].

Fig. 6 shows the deposition rate as a function of the discharge power for two different electrode distances and two excitation frequencies (13.56 MHz and 40.68 MHz). Each point corresponds to an optimized solar cell with respect to the transition between  $\mu$ c-Si:H and a-Si:H growth. An increase of deposition rate is observed by increasing the discharge power and/or by a reduction of the gap between electrode and substrate for both frequencies.

In further experiments we optimized the cell process for 40.68 MHz, achieved good homogeneity over the inner 26x26 cm<sup>2</sup> of our 30x30 cm<sup>2</sup> substrate size, and could enhance the deposition rate up to 2 nm/s. These experiments are described in a related paper [35]. Fig. 7 summarises the status of this work by highlighting the J-V-curves and corresponding photovoltaic parameters.



**Figure 6:** Deposition rate for  $\mu$ c-Si:H solar cells deposited with 13.56 MHz and 40.68 MHz as a function of the discharge power for two electrode distances.



**Figure 7:** J-V (current density – voltage) curves and corresponding photovoltaic parameters of illuminated  $\mu$ c-Si:H solar cells prepared at high deposition rates.

The i-layer thickness of these  $\mu\text{-Si:H}$  cells was between 1 and 2  $\mu\text{m}$ . At power densities below 1  $\text{W}/\text{cm}^2$  efficiencies of 9.4 % and 9.2 % were obtained at deposition rates of 0.5  $\text{nm/s}$  (13.56 MHz regime) and 1  $\text{nm/s}$  (40.68 MHz), respectively. At higher power densities and 40.68 MHz excitation still an efficiency of 7.9 % was maintained at a deposition rate of 2  $\text{nm/s}$ .

### 5.3 Up-scaling of the PECVD process to large areas

Different approaches are followed world wide to make the mass production of  $\mu\text{-Si:H}$  an economic reality. Due to the small process window for highly efficient  $\mu\text{-Si:H}$  cells, up-scaling is a very challenging task. In case of VHF excitation, problems concerning the process up-scaling towards square meter size like standing waves, skin effect or voltage uniformity may hinder homogeneous film growth (see [30,31] and references therein). To overcome these restrictions creative techniques have been developed, e.g. dielectric lenses [36], VHF line sources [37], ladder shaped electrodes [7] or ladder shaped antennas, which are implemented between two substrates facing each other [8]. Meanwhile, the question of the optimum excitation frequency with respect to deposition rate, cell performance, scalability to large areas and, finally, lowest production cost is still open. Moreover, the considered production concepts comprise high throughput batch reactors [9], in-line systems, which handle one or several substrates in parallel [8,10] and cluster type production machines [7].

The up-scaling of  $\mu\text{-Si:H}$  deposition to 1  $\text{m}^2$  was taken up by Applied Films GmbH & Co. KG in close cooperation with the IPV. For this purpose a planar PECVD plasma source with  $\sim 1 \text{ m}^2$  size has been implemented in a large area lab-coater [10,38]. Device quality  $\mu\text{-Si:H}$  cells with high efficiency as well as very good homogeneity of a-Si:H and  $\mu\text{-Si:H}$  films on large areas have been achieved [38]. First a-Si:H/ $\mu\text{-Si:H}$  tandem junction cells and modules were prepared on commercially available  $\text{SnO}_2\text{:F}$  coated glass yielding 11 % initial cell and above 10 % aperture area module efficiency. The described square meter process was operated at 13.56 MHz excitation frequency, yielding deposition rates close to 0.5  $\text{nm/s}$ . As next step, an enhancement of the deposition rate to  $\geq 1 \text{ nm/s}$  will be pursued by applying 40.68 MHz excitation frequency (see section 5.2). Further details and also first cost calculations are given in [38], which show the potential of the latter approach to provide a cost effective tool for mass production.

## 6 SUMMARY

Highly efficient thin-film solar modules were developed on glass substrates at low process temperatures using solely plasma assisted deposition techniques. A stable aperture area module efficiency of 10.1 % was obtained for an a-Si:H/ $\mu\text{-Si:H}$  module on  $10 \times 10 \text{ cm}^2$  substrate size.  $\text{ZnO:Al}$  films prepared by magnetron sputtering and post deposition wet chemical etching were applied as front TCO with excellent light scattering properties for silicon thin-film solar cells. This technique is compatible with high-rate sputtering from cost effective metallic targets. For optimised surface-textured  $\text{ZnO}$  films we obtained short-circuit current densities as high as 24.3  $\text{mA}/\text{cm}^2$  and 26.8  $\text{mA}/\text{cm}^2$  for

an i-layer thickness of 1.0 and 2.0  $\mu\text{m}$ , respectively, showing the potential for further improvements in  $\mu\text{-Si:H}$  solar cell efficiency. We developed  $\mu\text{-Si:H}$  i-layers by plasma-enhanced chemical vapour deposition (PECVD) on  $30 \times 30 \text{ cm}^2$  substrate size using 13.56 MHz and 40.68 MHz excitation frequency, high deposition pressures and high RF-powers. These conditions provide sufficiently “soft” deposition for the growth of high quality  $\mu\text{-Si:H}$  material and yield high deposition rates. Efficiencies of 9.4 %, 9.2 %, and 7.9 % were achieved for  $\mu\text{-Si:H}$  solar cells prepared at deposition rates of 0.5  $\text{nm/s}$  (13.56 MHz regime), 1  $\text{nm/s}$  (40.68 MHz), and 2  $\text{nm/s}$  (40.68 MHz), respectively.

Long term process stability and process reproducibility are prerequisites for a mass production of  $\mu\text{-Si:H}$  based solar modules. We have shown that even after rather long term operation reproducibly high module efficiencies can be obtained in a  $\mu\text{-Si:H}$  deposition regime of high pressure and high power. However, a careful optimisation of the  $\mu\text{-Si:H}$  process in a narrow deposition regime is required.

Subsequent up-scaling to  $\sim 1 \text{ m}^2$  coating area was performed together with the equipment manufacturer Applied Films using 13.56 MHz excitation frequency. Several a-Si:H/ $\mu\text{-Si:H}$  tandem modules, co-deposited on  $10 \times 10 \text{ cm}^2$  and  $30 \times 30 \text{ cm}^2$  substrates in a large area reactor, yielded initial aperture area efficiencies exceeding 10 %.

## ACKNOWLEDGEMENTS

The authors thank W. Appenzeller, H.-P. Bochem, S. Haas, R. Schmitz, J. Kirchhoff, B. Sehrbrock, H. Siekmann, W. Reetz, and Ch. Zahren from Forschungszentrum Juelich GmbH for their contributions to this work. Financial support by the BMU (contract 0329923A) is gratefully acknowledged. We thank Applied Films GmbH & Co. KG and RWE SCHOTT Solar GmbH for financial support and helpful discussions.

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