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FAST GROWTH OF MICROCRYSTALLINE SILICON SOLAR CELLS ON LP-CVD ZNO IN INDUSTRIAL KAI PECVD REACTORS

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ABSTRACT: We report in this paper on the latest research results of microcrystalline (μc-Si:H) silicon solar cells fabricated in a commercial Oerlikon Solar (former UNAXIS) KAI-S single-chamber PECVD reactor (substrate size up to 35 cm x 45 cm) driven at an excitation frequency of 40.68 MHz. The cell structure consists of a stack of glass/ front-TCO / p-i-n μc-Si:H solar cell / back-contact. Our "in-house" boron-doped ZnO (zinc oxide), prepared by LP-CVD (low-pressure chemical vapour deposition) is implemented for the front and the back-side transparent conductive contacts (TCO) of the devices. Homogeneous solar-grade films can be deposited at a rate up to 18Å/sec The Raman crystallinity of these films is of 62% in the center as well as on the border of the whole substrate area (35x45cm^2). By suitable optimisation of the device fabrication process we are able to make μc-Si:H solar cells with conversion efficiencies of 8.4% in a 'true' single-chamber process. The i-layer is 1.5μm thick and is deposited at 0.7 nm/sec on LP-CVD ZnO. The back ZnO contact is coated with white paint. Finally, we show that μc-Si:H solar cells with 6% efficiency can be grown, using so called 'low-flow' silane plasma in a depletion regime at a rate of 1 nm/s.

Keywords: Micro Crystalline Si, PECVD, Thin Film, Devices

1 INTRODUCTION

We report on an industrial research project at the University of Neuchâtel which had started in March 2004. The scope of this project is twofold: the first task is to upscale the fabrication process for hydrogenated microcrystalline silicon (μc-Si:H) solar cells from laboratory small area deposition (100 cm^2) up to a surface of 14000 cm^2 (the latter corresponds to the size of industrial deposition equipment sold by Oerlikon Solar [1]). The second task of the present project is to increase the deposition rate for the fabrication of solar-grade μc-Si:H absorber layers and devices. High-rate deposition is necessary to achieve short fabrication times for the relatively thick μc-Si:H absorber layer in a micromorph tandem solar cell: for comparison, in such a tandem structure the amorphous top cell is thinner than 0.3 μm, contrary to the microcrystalline bottom cell which should typically be 1 to 2 μm thick. To be fully "industry compatible" the processing for all doped and intrinsic microcrystalline silicon layers prepared at high deposition rates has to be executed within a single-chamber PE-CVD reactor.

As was already shown in a preceding research project [2], Oerlikon Solar KAI PE-CVD reactors developed for active-matrix LCD technology possess a high potential for cost-effective manufacturing of thin-film silicon solar cells based on amorphous silicon. The development of a fabrication process for microcrystalline silicon solar cells on the same industrial reactor is the aim of this present project.

Thereby, specific issues relating to the preparation of microcrystalline devices had to be addressed, using both the small KAI-S reactor at IMT Neuchâtel, and the larger area KAI reactors at Oerlikon Solar. A successful conclusion of the project will allow for cost-effective production of micromorph modules in the future.

2 EXPERIMENTAL

2.1 Organization of the work

The work was divided into two parts: In a first time the **parameter space** for deposition conditions (total gas flow, pressure, plasma power, silane concentration) was scanned. Some hardware modifications had to be implemented to achieve this milestone. Then, in a second time of this project we focused entirely on **device optimization** using thereby a single-chamber process (i.e. the sample remains in the chamber during the whole processing cycle).

2.2 Description of the system

The full device manufacturing procedure can be divided into three parts: 1) the front-contact consists of IMT's modified LP-CVD (Low Pressure Chemical Vapour Deposition) process [3] in order to fabricate growth-textured zinc oxide films (ZnO) for enhanced light-trapping on AF45 Schott glass. The sheet resistance of the resulting front-contact layers is below 10Ω/$\square$. Then, 2) these ZnO coated glass substrates are introduced to the KAI-S reactor for deposition of the μc-Si:H solar cell. The deposition system used for this work at IMT is an adapted version of the industrial KAI-S reactor commercialized by Oerlikon Solar. It is a parallel-plate capacitively-coupled reactor, with substrate dimensions of 35 cm x 45 cm (width x length). The Plasma-Enhanced Chemical Vapour Deposition (PE-CVD) is used as deposition method, with an excitation frequency of 40.86 MHz. This KAI-S reactor has been equipped with a load-lock chamber which allows for shorter deposition cycles and for better vacuum conditions in the deposition

chamber. The R&D groups of Oerlikon Solar perform the fabrication of devices both in KAI-M (52 x 41 cm^2) and KAI 1200 (1.4 m^2 substrate size) reactors. 3) The back-contact ZnO is then deposited again by LP-CVD onto the silicon film, sub-cells of approximately 0.25 cm^2 are patterned and current-voltage (I-V) curves are measured for each sub-cell under a WACOM AM1.5 solar simulator. The short-circuit current density is calculated by forming the product of the external quantum efficiency (EQE) measurement and the solar spectrum (expressed by the spectral density of photon flux) and an integration over a range of 350nm to 1000nm.

The thickness of the μc-Si:H devices is determined by a TENCOR alphastep 200 profilometer and the deposition rate can then be calculated. A commercial Renishaw Raman imaging microscope (System 2000) with an Argon laser excitation beam (λ = 514nm), is used to evaluate the crystalline fraction of the films and solar cells. The Raman crystallinity was calculated as

$\Phi_C = (I_{520}+I_{510})/(I_{520}+I_{510}+I_{480})$ where I_{520}, I_{510}, I_{480} is the total integrated area under the Gaussian centered at 520, 510 and 480 cm^{-1} respectively [4].

3   RESULTS

3.1 Layer optimization

The first task was to explore the working regimes for μc-Si:H film deposition with an equipment which was never used before in such conditions. Since the parameter space necessary for solar grade AND fast μc-Si:H thin-film deposition is known to extend into more extreme conditions than usually applicable for the fabrication of amorphous silicon solar cells, i.e. one needs high plasma power, high working pressure and high deposition gas flow as it is suggested by recent work of other research groups in Germany and Japan [5, 6].
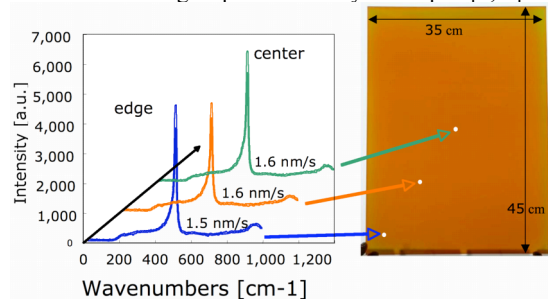
Figure 1: Raman crystallinity of microcrystalline silicon films grown in the KAI-S reactor at IMT at deposition rates exceeding 1.5 nm/s. The Raman probe (λ=514 nm) is taken from the center to the edge of the substrate area (35x45cm^2. The film has in the center a thickness of 0.96 μm.

The systematic exploration of the parameter space for depositing high-rate intrinsic μc-Si:H absorber films of device quality (=solar grade films) was investigated by variation of key parameters, like deposition pressure, silane concentration and injected plasma power. This task required a very intensive work effort given the high level of complexity of the microcrystalline growth process. Fig. 1 reflects the best results on the i-layer optimization obtained in the KAI-S reactor, in terms of increasing deposition rate: it shows the Raman spectra (incident light at λ=514 nm) of a microcrystalline layer deposited on a 35x45 cm^2 substrate as well as the deposition rates obtained. Homogeneous and high crystallinity of μc-Si:H

layers over the whole substrate area is achieved at a rate exceeding 1.5 nm/sec, and shows that high-quality μc-Si:H can be prepared in KAI reactors.
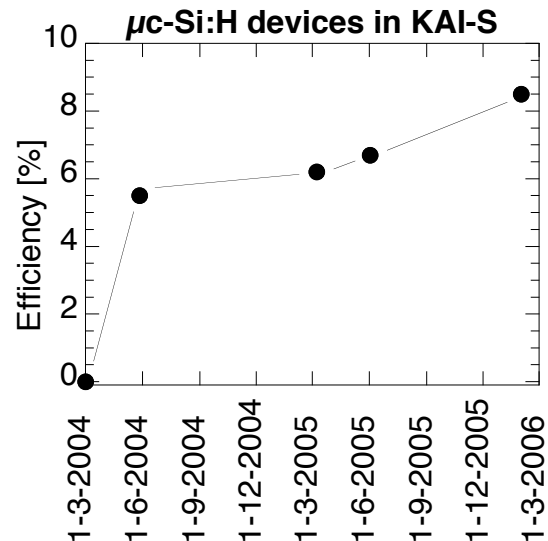
Figure 2: Conversion efficiency of μc-Si:H p-i-n solar cells fabricated in the KAI-S versus project time.

3.2 Device optimization

After the achievements on the side of intrinsic high-rate layer optimization and decent doped layers, first μc-Si:H devices were tested. In Fig. 2, the progress on conversion efficiency is plotted versus the project time period, with a best cell at 8.4% efficiency and a V_{oc}-value of 5134mV, FF=73% and J_{sc}=22.4mA/cm^2. Note, that for this final result of 8.4% (see I-V curve in Fig. 3), ZnO of the LP-CVD process at IMT is utilized as front contact (after application of a surface treatment [7]) and as back contact. The μc-Si:H device was fabricated in a 'true' single-chamber process, i.e. the substrate remains in the plasma zone during the whole deposition process without using the load-lock. The deposition rate of the absorber layer was 0.7 nm/s.

A final emphasis has been put on the homogeneity of μc-Si:H devices. Five substrates have been distributed over the whole electrode area of 35x45 cm^2 for this purpose. The resulting 75 values for V_{oc} and fill factor reach in average 513mV and 67% respectively and have both a relative standard deviation of only 2.2%.

3.3 Growth of μc-Si:H cells at very low gas flow

Many recent publications of high-rate growth of μc-Si:H films focused on the so-called 'high-pressure depletion' (HPD) regime [5, 6]. Two typical conditions for this regime are high feedstock gas flux (several *slm*) and high plasma power (injected plasma power density > *1W/cm^2*). Such conditions can be an economic drawback for the commercial production of μc-Si:H solar cells. We showed in earlier studies [8] the feasibility to fabricate first μc-Si:H devices in the so-called 'closed-chamber VHF' regime, where extremely low feedstock gas flow (some *sccm*) and moderate plasma power (< 0.5 W/cm^2) are utilized. Later on, state-of-the-art μc-Si:H devices could be presented [9, 10]. Recently these closed-chamber conditions have been analyzed further using the process conditions of CRPP [11,12] on an industrial KAI plasma reactor.

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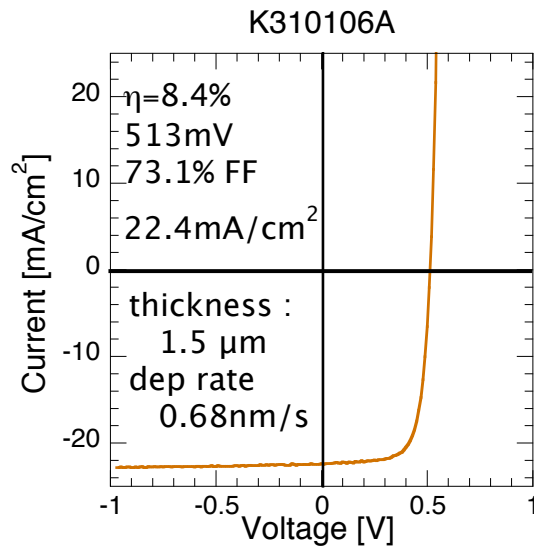
Figure 3: IV curve under AM1.5 illumination of the state-of-the-art μc-Si:H solar cell fabricated in a 'true' single-chamber process in the KAI-S reactor.

In our experiments, we found that very low flow of silane gas and hydrogen gas (typically 50 to 100% silane concentration) together with moderate plasma power are indeed suitable to fabricate μc-Si:H devices: The best devices so far reach a conversion efficiency of **6%** (V_{oc} = 492mV, FF = 73%, J_{sc} = 17 mA/cm^2) and are fabricated for the first time in a KAI-S plasma reactor under 'low-flow' conditions at a deposition rate of 1 nm/s. Such a 'low-flow' condition is similar to the former 'closed-chamber VHF' deposition technique since a completely depleted silane plasma is maintained steadily up to the desired absorber layer thickness. The minimum necessary silane gas flow is calculated based on the desired deposition rate and under the assumption of maximum gas consumption within the plasma [11, 12]; plasma excitation frequency is the same as in conventional gas flow conditions at 40MHz. Further optimization will be necessary in order to reach the state-of-the-art conversion efficiency of 8.4%. Note that the fabrication of an absorber layer of 1.2μm thickness in the 'low-flow' regime needs more than 10 times less gas (hydrogen and silane) than the fabrication of a 'conventional' 1.5μm thick absorber layer. Pure silane plasma can be used for μc-Si:H device growth in large area reactors because, when the silane is strongly depleted, the plasma composition is almost entirely hydrogen [11,12]. The hydrogen dilution in the plasma itself is consequently much higher than the hydrogen dilution estimated from the input flow rates.

4 CONCLUSIONS

The utilization of the KAI-S type PECVD reactor as now equipped at IMT has been verified for industrial photovoltaics processing: The system initially constructed for the flat-panel processing industry could be transformed into a tool for the thin-film silicon solar cell processing industry aiming at the micromorph tandem cell concept. This has been achieved by some hardware modifications and by intensive parameter scans in order to reach solar grade microcrystalline silicon films at high deposition rates: the best μc–Si:H solar cell

device reaches a conversion efficiency of 8.4% at a deposition rate of 0.7 nm/s in a "true" single-chamber process. Several high-efficiency micromorph modules could be prepared by Oerlikon Solar. One additional process window for μc-Si:H device fabrication has been identified: there is not only the 'conventional' high-power & high gaz-flow regime for fast device grade μc-Si:H material fabrication, there is also an interesting fully depleted 'low-flow' regime where sofar 6% efficiency μc-Si:H devices could be fabricated using 10 times less feedstock gaz.

A final emphasis has been set on the homogeneity of the μc-Si:H devices; this last milestone could be satisfactorily achieved. This work will prepare the way for the low-cost fabrication, at an industrial level, of state-of-the-art "micromorph" tandem modules.

5 ACKNOWLEDGMENTS

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