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Microcrystalline/micromorph silicon thin-film solar cells prepared by VHF-GD technique

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Abstract

Hydrogenated microcrystalline silicon prepared at low temperatures by the glow discharge technique is examined here with respect to its role as a new thin-film photovoltaic absorber material. XRD and TEM characterisations reveal that microcrystalline silicon is a semiconductor with a very complex morphology. Microcrystalline p–i–n cells with open-circuit voltages of up to 560–580 mV could be prepared. "Micromorph" tandem solar cells show under outdoor conditions higher short-circuit currents due to the enhanced blue spectra of real sun light and therefore higher efficiencies than under AM1.5 solar simulator conditions. Furthermore, a weak air mass dependence of the short-circuit current density could be observed for such micromorph tandem solar cells. By applying the monolithic series connection based on laser patterning a first micromorph mini-module (total area of 23.6 cm^2) with 9% cell conversion efficiency could be fabricated.

Keywords: Hydrogenated microcrystalline silicon; Thin-film crystalline silicon; VHF-GD; Micromorph concept; Monolithic series connection

1. Introduction

In order to make photovoltaics a competitive energy source in the future, it is imperative to adopt an approach that enables the fabrication of low-cost solar cells. Ways to meet low-cost and high efficiency can possibly be found within the different existing concepts of thin-film solar cells [1,2]; thereby the "technically safe" but expensive wafer with its phenomenally long diffusion length of hundreds of

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micrometers has to be abandoned. Special efforts have, therefore, to be undertaken to tailor the required properties of the thin-film absorber materials.

A promising technique to obtain thin-film solar cell devices is the low-temperature PECVD deposition method. Using this technique, only amorphous silicon-based solar cells are produced today at the scale of 1–10 MW-manufacturing units. In 1994 our group demonstrated, using the low-temperature PECVD, that hydrogenated microcrystalline silicon (μc-Si:H) can also be an excellent photovoltaic absorber. This new type of thin-film crystalline silicon (energy gap of 1.1 eV [3,4]) can advantageously be combined with the well-approved amorphous silicon technology to form so-called *micromorph* (*micro*crystalline/a*morph*ous) tandem solar cells [5–15]. This micromorph cell is at present considered to be one of the most promising thin-film solar cell concepts that may possibly be able to meet the challenge for a low-cost high-efficiency solar cell for widespread terrestrial use [13–15]. It has to be noted that such microcrystalline silicon is the first form of non-wafer-based crystalline silicon that can be directly combined with amorphous silicon to conciliate the low-cost and high-efficiency requirements for a large-area photovoltaic technology. Moreover, the low-temperature plasma process involved allows for high flexibility in the use of cheap substrates (e.g. glass, metals, polymers, etc.) as well as for the deposition of both p–i–n and n–i–p cell structures. Thus, micromorph tandem solar cells in both configurations could successfully be fabricated showing, thereby, stabilised efficiencies in the range of 11–12% [8,14,15].

In spite of the surprising results of μc-Si:H single-junction solar cells achieved so far, there is a lack of understanding with respect to the basic properties of intrinsic μc-Si:H when used as absorber material, as well as with respect to the mode of functioning of the whole solar cell device [3,16]. In this paper we show that the morphology on so-called "microcrystalline silicon", grown by very high-frequency glow discharge (VHF-GD) technology, is extremely complex. The different features of the grains suggest that one should rather talk of microcrystalline silicon semiconductors in the plural form as sofar no standard type of μc-Si:H can be made out.

Micromorph tandem cells were characterised under clear outdoor conditions and compared with the measurements done by our laboratory sun simulator.

A key issue for every thin-film photovoltaics concept is the application of the monolithic series connection of the solar cells. In this paper we present this proof of concept by giving results on our first laser-patterned "micromorph mini-modules".

2. Results and discussion

2.1. Morphology of VHF-GD microcrystalline silicon

Microcrystalline silicon can be prepared using a wide range of deposition parameters. Its growth regime is, in principle, limited by the transition to the amorphous growth. Here, we present a series of undoped microcrystalline samples prepared (on Schott AF45 glass) by varying only the silane dilution parameter, keeping all other deposition parameters constant [17].

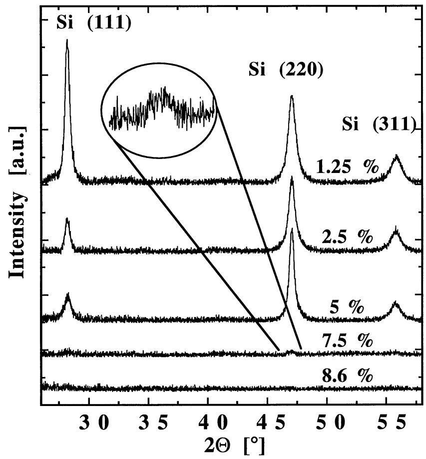

Fig. 1. X-ray diffraction patterns of a dilution series of μc-Si:H samples deposited on Schott AF45 glass substrates [17].

X-ray diffraction (*XRD*) *measurements* in Fig. 1 reveal a crystallographic texture in the (2 2 0) direction. This preferential growth, however, is strongly dependent on the dilution ratio parameter of the deposition and is most pronounced around a dilution of 2.5%. At low silane concentrations of 1.25% this preferential texture is reduced, but still present. The XRD pattern of Fig. 1 shows that in contrast with the case of amorphous silicon in microcrystalline silicon the internal crystallographic structure is heterogeneous and depends sensitively on the dilution parameter.

In order to obtain more insight into the internal microstructure, *transmission electron microscopy* (*TEM*) *characterisations* [18] have been undertaken on this undoped μc-Si:H film series of Fig. 1. In Figs. 2–4 cross-section micrographs of three samples from the dilution film series are shown. As can clearly be seen, the features of the grains are quite different for each layer: the 1.25% diluted film in Fig. 2 shows up with 750 nm long hexagonal (20 nm diameter) columnar grains, resulting in an extremely rough surface. A discontinuity of the microstructure is observed in the first 40 nm close to the glass–silicon interface (upper left corner in Fig. 2). This initial zone of the layer consists of small isotropic grains, indicating that the growth mechanism during deposition has only reached steady-state conditions after a certain thickness/time. For the 2.5% diluted sample, as shown in Fig. 3, one can observe large bunches (200 nm diameter) which consist of several small grains (20–30 nm diameter) resulting also in a rough dendritic surface. Beside these “cauliflower”-like bunches one can identify some large (50 nm diameter) leaf-like grains with a central stacking fault axis. Such leaf-like grains are similar to those found in thermally recrystallised a-Si:H material as used for current TFT applications [19,20]. This sample reveals that the VHF-GD technique may allow to deposit directly such TFT-like material at low temperatures (200–250°C); thus, the substrate choice is free and diffusion contaminations of underlying structures are considerably reduced. Although an X-ray peak is

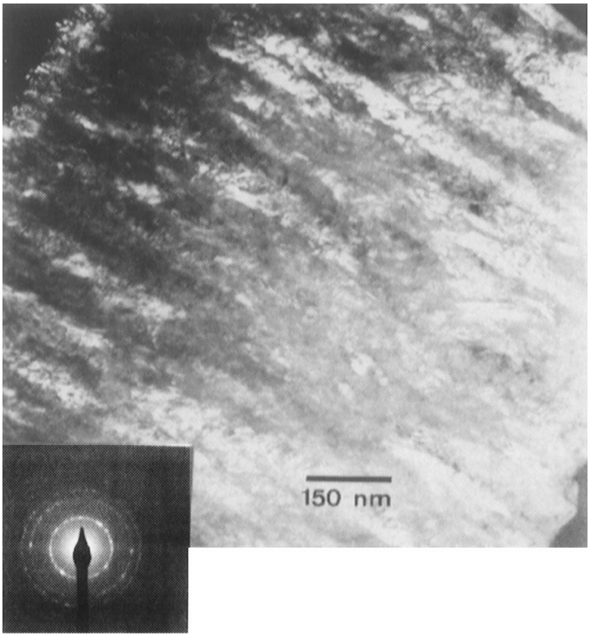

Fig. 2. Cross-section dark-field micrograph of a sample prepared at a silane concentration of 1.25% ($[SiH_4]/[SiH_4 + H_2]$). The selected diffraction pattern (in inset) with its dotty rings is indicative of the microcrystalline nature of the layer [18].

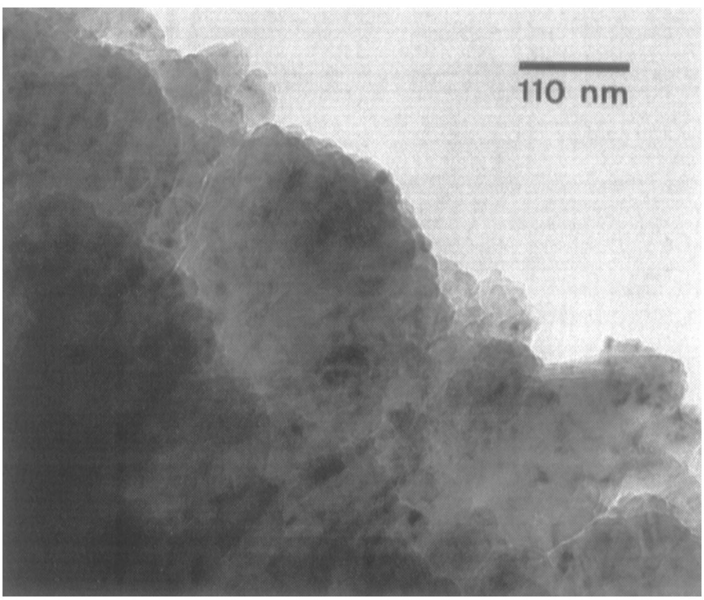

Fig. 3. Cross-section bright-field micrograph of a μc-Si:H layer grown at a concentration of 2.5% ($[SiH_4]/[SiH_4 + H_2]$) silane in hydrogen [18].

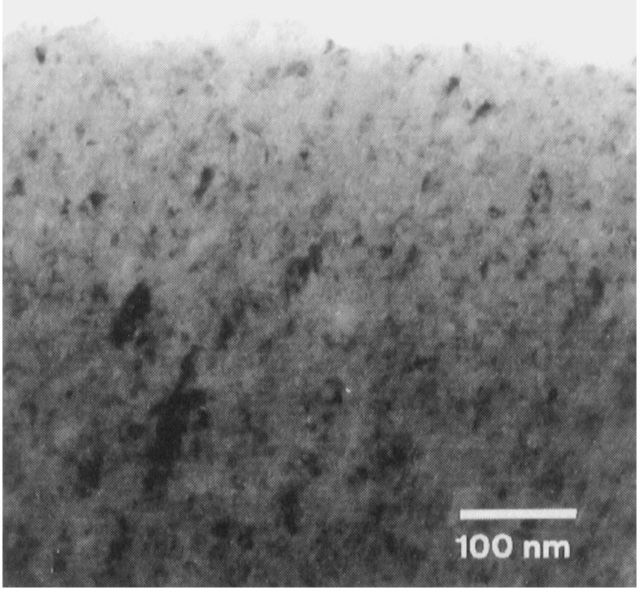

Fig. 4. Side-view bright-field micrograph of a sample prepared at a silane concentration of 7.5% ($[SiH_4]/[SiH_4 + H_2]$). The shapes of 30–50 nm elongated crystallites can be observed [18].

barely observed for the 7.5% diluted sample, one can observe small (10 nm diameter) elongated nanograins of 30 nm length in the TEM micrograph of Fig. 4. Image analysis of a plane-view micrograph indicates that about 10% of the material volume is crystalline [18].

The above TEM results are partly supported by another, independent TEM dilution series study carried out by the Jülich group on n-type μc-Si:H silicon [21]; in the latter study columnar microstructures and elongated nanocrystallites have also been observed, however, without a marked preferential (2 2 0) orientation, as in our case. This may be due to the use of different substrates or due to the difference in deposition conditions.

These structural results reveal that in contrast to amorphous silicon the term of the so-called "microcrystalline silicon" is not related to a single typical "standard" material but has to be associated with a large variety of different μc-Si:H materials, all with different grain morphologies and crystallographic textures. Thus, based on the huge variety of internal microstructures present in microcrystalline silicon, one may expect substantial differences in the corresponding electronic and optical properties, especially when these materials are incorporated into *microcrystalline solar cell devices*. For example, as the electronic transport in a μc-Si:H solar cell is parallel to the film growth, the features of the grains (size, form, etc.) as seen in this direction and the average number of internal grain boundaries encountered on such a transport path may have a certain influence on solar cell performance.

In conclusion, microcrystalline silicon has to be considered as a thin-film semiconductor with an impressive richness of complex internal features. Such a rich variety of μc-Si:H materials implies that different properties will result. All these structural properties are sensitively influenced or can even be controlled by the applied deposition parameters (e.g. deposition technique, temperature, VHF-power, pressure, dilution level, substrate; just to mention a few).

2.2. *Microstructure and solar cell performance*

What impact have such different microstructures on cell performance? A full p–i–n solar cell device, due to the different layers involved, is much more complex and delicate to interpret than isolated μc-Si:H thin-films alone. The performance of an individual μc-Si:H layer within a p–i–n device may be affected not only by its own complex structure but also by the electronic and optical properties of other layers, and even further by contact problems. Hence, it is not straightforward to find a correlation between internal structure and device properties. A decisive influence on the ongrowing structure certainly originates also from the first initial doped "seeding" layer; in the case of a p–i–n structure, this is the p-window layer. As evidenced in the work of the Kaneka group, the crystallinity condition of this initial "seeding" layer is fundamental for the quality of the resulting solar cell [22]. Doped μc-Si:H p-type layers can, however, be prepared under various conditions [23,24] and their exact properties depend on the substrates used. This means that the films of Figs. 2–3 may have different structural features when deposited under the same conditions, but on a glass/TCO substrate covered by a p-type doped μc-Si:H rather than on bare glass substrates (as used for the samples analysed by TEM and described above). This is especially decisive, in the case of the p–i–n structure, because the properties of the photovoltaically important p/i interface are formed at the beginning of the nucleation zone; a large part of the light is converted in this zone; thus, this zone becomes an essential bottleneck for the performance of the whole solar cell, even if the ongrowing bulk absorber has an ideal structure with respect to electronic carrier transport.

At the present stage we are not able to give a full picture for the microstructure-related behaviour of our μc-Si:H cells. Various cell parameters such as light-trapping, the thicknesses, structural inhomogeneities vs. thickness were not always the same, masking, thus the effects due to the intrinsic absorber layer.

However, one may even so observe a certain confirmed tendency for the *open-circuit voltage of such single-junction μc-Si:H cells*. In Fig. 5, the V_{oc} values of a large number of p–i–n cells deposited under a wide range of different deposition conditions are plotted in function of the silane dilution parameter of the incorporated intrinsic μc-Si:H absorber layer. All cells, except for the amorphous one (deposited at a dilution of 9.5%), show a clearly enhanced infrared response in the region above 750 nm and are therefore classified as being microcrystalline. There is a general trend for a V_{oc} increase with decreasing silane dilution as one approaches the transition to amorphous silicon, which is located somewhere between 7% and 8%. This is somehow astonishing: the sample deposited at the highest dilution of 1.25% is a sample with a high crystallinity consisting of long columnar grains (up to 750 nm). Such structures were up to now believed to be best suited for electronic transport (in the direction perpendicular to the substrate), since carriers have only a small number of

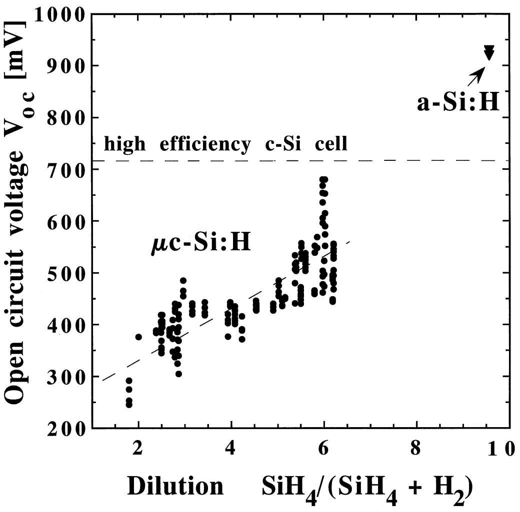

Fig. 5. V_{oc} values for a large number of different p–i–n μc-Si:H solar cells fabricated at IMT Neuchâtel in function of the silane dilution parameter ($[SiH_4]/[SiH_4 + H_2]$).

grain-boundaries to overcome. Surprisingly, such layers incorporated into solar cells show the lowest V_{oc} values. It is somehow paradoxical that smaller grains and, thus, more grain boundaries in the material lead to higher V_{oc} values. From the observed nucleation zone in Fig. 2 one may speculate that the p/i interface becomes highly disordered at high dilutions, this being responsible for a reduced V_{oc}. An optimisation of this initial growth zone towards large grains starting immediately at the substrate surface and passing through the entire cell thickness may possibly lead to solar cells with higher values of V_{oc}. In order to obtain more insight into the complex structure of the grains and its relation to the cell properties, further broad structure related investigations have to be performed on different type of μc-Si:H cells.

2.3. Recent μc-Si:H p–i–n solar cells

The efficiency of the micromorph tandem cell is directly linked to the open-circuit voltage of the microcrystalline silicon bottom cell. Our first microcrystalline cells suffered from rather low open-circuit voltages of only 350–400 mV [3,5]. Higher V_{oc} values were in the beginning always related to low fill factors. Recent results, however, on μc-Si:H p–i–n cells reveal that open-circuit voltages of up to 530 mV with simultaneously high FF-values of 70% are feasible; thereby an efficiency of 8.5% for a single-junction μc-Si:H cell could be obtained [9,10]. In the meantime, further efforts have been undertaken with the emphasis on improving the FF of high-V_{oc} cells. Note that light-trapping properties, e.g. the transparency of the p-layer and the quality of the back contact reflector, have sofar not been considered in these preliminary cells. The thickness of the absorber layer was kept between 1 and 2 μm in order to reduce the deposition time. The main goal of this study was to assess the possibility of

Table 1
Recent microcrystalline p–i–n solar cells with improved V_{oc} and FF

η (%)	8.5	7.3	5.7	6.3	6	5.6	4.1
J_{sc} (mA/cm^2)	22.9	20.2	15.9	17.5	16	14.8	15.5
FF (%)	69.8	66.7	65.2	64	66	64.5	38.7
V_{oc} (mV)	531	539	550	560	569	582	689

fabricating cells having both a high V_{oc} and a high FF. Recent results of small-area p–i–n cells (0.1–0.2 cm^2) are given in Table 1.

Obviously, from these preliminary microcrystalline silicon cells can be seen, high V_{oc} values are not inconsistent with high FF. Hence, it seems that the μc-Si:H material has the potential for cells with open-circuit voltages of 560–580 mV and reasonably high fill factors. Further extensive work and further optimisation, especially with respect to light-trapping properties, will be necessary in order to tap the full efficiency potential of the μc-Si:H solar cell device.

2.4. *Proof of concept: Micromorph "mini-module" fabricated by laser patterning*

An important question for any thin-film solar cell is the applicability of the monolithic electrical series connection method for individual cells. In contrast with wafer-based silicon solar cells, thin-film technologies allow for the direct deposition of the solar cell absorber on large substrate areas. Hereby, the output characteristics of such a substrate unit can be adapted, by the help of structuring techniques and parallel/series connection of the cell segments, to form any desired compromise between a high-current module and a high-voltage module.

In amorphous silicon p–i–n-based module technology such monolithic series connection is well established and uses the laser-scribing patterning. The structuring of the front TCO and of the back contact and of the absorber material is realised by adapted ablative laser light pulses (Nd : YAG, 1064 and 532 nm). Microcrystalline or micromorph tandem cells are, however, much thicker than a-Si:H-based cells; thus, the question remains if this laser-scribing technique is also applicable on 2–3 μm thick silicon-absorber films without damaging the cells.

Applying all three scribe lines, for the front TCO (1064 nm) and back contact (532 nm) and for the micromorph tandem cell (532 nm), we succeeded in the fabrication of our first micromorph "mini-module" as shown in Fig. 6 of a total surface area of 23.6 cm^2. It consists of six cell segments, each one of an area of 3.8 cm^2. The I–V characteristic results in a total module efficiency of 9.0% (V_{oc} of 7.95 V, J_{sc} of 1.643 mA/cm^2, FF of 69.0%). No light-trapping schemes were sofar applied for these first modules, which result in a relative low short-circuit current density. The fact that the total module voltage (7.95 V) is almost equal to the sum of the single-segment V_{oc} values (6×1.33 V) and the resulting module fill factor of 69% indicate that such a laser-scribing patterning (as used for a-Si:H-based module manufacturing) is in principle also feasible for our thicker micromorph cells.

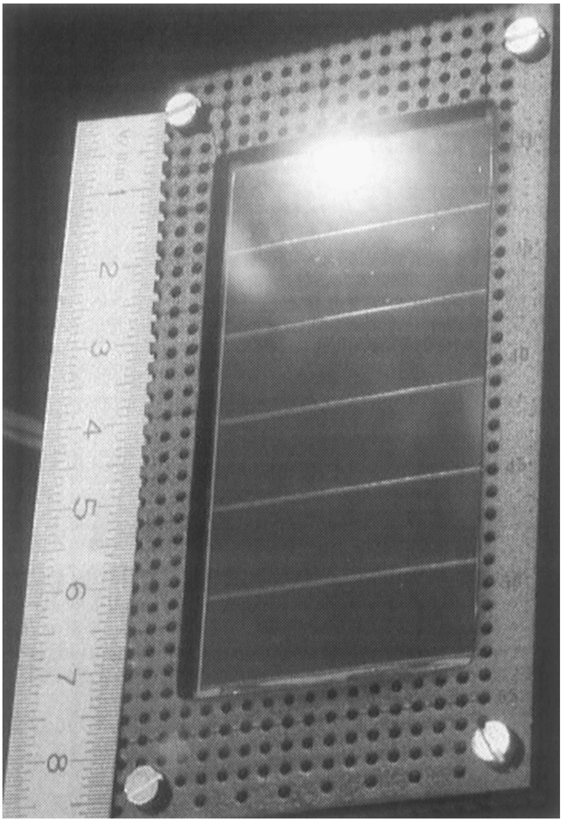

Fig. 6. Proof of the monolithic series connection concept: first IMT micromorph mini-module (23.6 cm^2) prepared by applying the laser-scribing technique.

These first modules indicate furthermore, that an up-scaling of the micromorph concept from sofar 1 cm^2 size cells to larger area cells is fully practicable. Homogeneity aspects are probably similar as in the amorphous silicon-based technology, which means that the manufacturing of large m^2-size area micromorph modules should in principle be possible.

2.5. Micromorph tandem cells under outdoor conditions

The short-circuit current density, J_{sc}, of tandem solar cells depends sensitively on the light spectrum used for the *I–V* characterisation. Especially, when semiconductor materials with large differences in the energy gap are involved, as is the case with micromorph tandem cells (amorphous top of 1.7 eV, microcrystalline bottom of 1.1 eV), then the determination of the exact J_{sc} value for AM1.5 conditions becomes critical. An artificial solar simulator has to simulate as best it can AM1.5 conditions over a wide wavelength range and this can only be achieved approximately in the

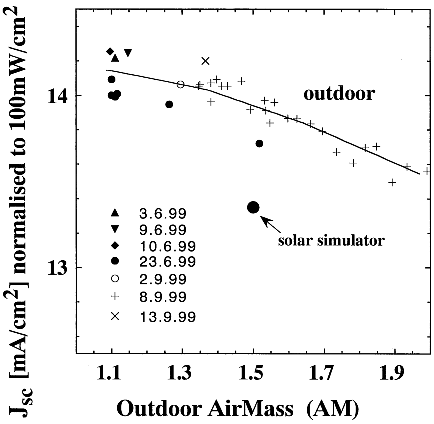

Fig. 7. Short-circuit current measurements of a 1 cm^2 micromorph tandem cell under outdoor and indoor illumination (two-source simulator, Wacom WXS-140S-10) normalised to 100 mW/cm^2. The light intensity was determined by a calibrated monocrystalline silicon solar cell. The air mass (AM) value was determined by measuring simply the incident angle of sunlight. Note that due to atmospheric variations (dust, humidity, temperature), outdoor conditions can always differ slightly from one another.

laboratory by using different lamp sources. In order to check the influence of the spectrum to the micromorph cells we performed outdoor measurements in Neuchâtel (northern latitude of 46°59.6′, 430 m above sea level) and compared them with indoor results obtained by our own solar simulator (Fig. 7). For the indoor measurements a two-source (xenon and halogen lamps) simulator (Wacom WXS-140S-10) was at our disposal. To get better statistical confidence, the outdoor measurements were performed on several days in the year. By that the influence of slightly different atmospheric conditions (dust, humidity) are better taken into account, thereby allowing also for a better judgment of the measurements. As test cells, micromorph tandems were used with well-defined patterned areas of 1 cm^2. Peripheral collection effects are, thus, excluded because the patterning of these devices were done in a precise way, by the laser-scribing technique. Relative spectral response measurements of the outdoor test cells indicate that the micromorph tandems are "top-cell limited", in the case of the cell of Fig. 7 the difference is approximately 8%. As reference detector we used a calibrated monocrystalline solar cell (of 4 cm^2 area, recently certified by ISE Freiburg), both for our solar simulator as well as for AM1.5 global conditions. The AM1.5 global calibration was used for the normalisation of the outdoor measurements to 100 mW/cm^2. A later check of the calibrated reference cell with a broadband independently calibrated thermopile (model CM21 from Kipp&Zonen) showed, under outdoor conditions, ranging from AM1.36 to AM2, an excellent agreement of less than 1% deviation in the outdoor light intensity measurement. The air mass value was determined by the inclination angle of the sun to the horizontal plane. During the outdoor measurements the temperature of the micromorph tandems was controlled by a Pt-100 sensor; the measurements were performed between 23°C and 30°C. Earlier temperature-dependent measurements have shown that the influence of the temperature on the short-circuit current of micromorph cells is very small (TC(J_{sc}) $\sim 7.7 \times 10^{-4}$ [9]); thus, we can conclude that under the above conditions we have a maximal temperature-related error of $\pm 0.06\,mA/cm^2$.

The comparison between outdoor and simulator measurements show clearly that micromorph cells reveal under realistic reporting conditions (outdoor) up to 0.6 – $0.8\,mA/cm^2$ higher short-circuit currents densities than under artificial AM1.5 illumination. The higher short-circuit currents under outdoor conditions indicate that the blue part of the clear sky spectra is enhanced leading thereby to a better performance of the micromorph tandem cells than under the indoor AM1.5 solar cell standard. The weak dependence on the outdoor air mass may be attributed to the relative large range of spectral sensitivity of both the top cell and of the bottom cell; this being due to the relative large difference in their bandgaps. The advantage of the high blue and green sensitivity (somehow adapted to the high-energy part of the sun spectrum) of amorphous silicon appears here to be ideally combined with the enhanced red and infrared response of the microcrystalline bottom cell when forming the micromorph tandem cell structure.

A few done outdoor *I–V* measurements, sofar investigated under air masses lower than 1.5, show furthermore that the FF and V_{oc} values of micromorph tandems are in principle not affected when compared with indoor AM1.5 *I–V* characteristics. Therefore, micromorph tandem cells may have under realistic conditions (shown here for a bright clear sky) a better performance than under standard AM1.5 illumination. Of course long-term outdoor field testing experiments of whole modules under different meteorological conditions is now necessary to allow for more detailed and precise information on the actual outdoor micromorph tandem cell behaviour.

3. Conclusions

Intrinsic microcrystalline silicon deposited at temperatures as low as 200–250°C by the VHF-GD method is now widely accepted as a new photovoltaically active material for use within p–i–n and n–i–p-type solar cells. Within this paper the authors have shown that the open-circuit voltage of microcrystalline p–i–n solar cells, which is directly related to the micromorph tandem cell efficiency, can be increased upto values in the range of 560–580 mV, whilst still obtaining reasonably high fill factors.

Microcrystalline silicon is not just an individual new semiconductor absorber for solar cells, but is a material which contains an incredible richness of different morphologies, as was revealed in this work by TEM and XRD studies. The resulting microstructures and morphologies depend strongly on the applied deposition parameters. Looking at very many different μc-Si:H cells fabricated at IMT Neuchâtel one notes that the value of V_{oc} generally increases with silane content in the silane/hydrogen feedgas mixture used.

The determination of the short-circuit current of tandem cells is, in general, critically dependent on the light spectrum used. Outdoor measurements under clear, cloudless sky conditions reveal that micromorph tandem cells have higher short-circuit currents and, hence, higher efficiencies under real sun illumination when compared to indoor AM1.5 solar simulator conditions.

A proof of concept for monolithic series connection of micromorph solar cells could be given: a first micromorph "mini-module" was fabricated. By the laser-scribing technique — as already well-established for the structuring of amorphous silicon solar cell modules — a micromorph tandem "mini-module" ($23.6\,cm^2$) with 9% total area cell efficiency could be obtained. This compatibility of the monolithic series connection method with the micromorph thin-film solar cell concept is an important issue when considering the direct fabrication of large-area modules, as is essential in reducing future manufacturing costs.

Acknowledgements

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