Baseline meets innovation: Technology transfer for high-efficiency thin-film Si and CIGS modules at PVcomB

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ABSTRACT
Thin-film PV modules are one of the most sustainable options for the generation of electricity, with low material consumption and short energy-payback times. Both of these factors are essential for paving the way towards a terawatt PV market. However, the cost-competitive production of PV modules has become extremely difficult, and module producers are facing huge challenges. A rapid technology transfer from research to industry is therefore required in order to introduce innovations for lower production costs and higher conversion efficiencies. At the Competence Centre Thin-Film- and Nanotechnology for Photovoltaics Berlin (PVcomB), founded by the Helmholtz-Zentrum Berlin (HZB) and the Technical University Berlin, two R&D lines for 30×30cm^2 modules based on thin-film silicon and copper indium gallium (di)selenide (CIGS) respectively are operated. Robust baseline processes on a high efficiency level, combined with advanced process and device analytics, have been established as a basis for the introduction and development of further innovative technology steps, and their transfer to industry.

PVcomB's mission

Over the last few years, thin-film (TF) PV has been successfully established on the PV market. Large-area production technologies and the use of substrates such as glass or foil are just two means of achieving low-cost production. Different material concepts – such as thin-film silicon, CIGS (compounds based on copper, indium, gallium, sulphur and/or selenium) and cadmium-telluride – are currently in mass production; they have already shown their potential and ability to realize low costs and high volumes in industrial production. But the current market crisis has considerably increased the pressure both on conventional PV production of crystalline Si modules and on TF technologies. The need for significant cost reductions and higher conversion efficiencies is greater than ever.

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The Competence Center Thin-Film- and Nanotechnology for Photovoltaics Berlin (PVcomB) expressly addresses these topics by bridging the gap between fundamental research and industrial application. PVcomB is therefore focusing on the following topics: 1) technology transfer from the lab to a cost-effective production line; 2) R&D in all aspects of production of thin-film PV modules; and 3) education and training of highly skilled, thin-film PV professionals.

To this end, PVcomB has set up in the last three years two R&D TF PV reference lines (TF Si and CIGS) on an intermediate module size of 30×30cm^2. The whole process chain from glass-substrate washing to the encapsulation of fully processed solar modules is covered, complemented by advanced analytical tools for in situ and ex situ process analytics and high-level device characterization. A truly unique feature of PVcomB's reference lines is that both technologies (TF Si and CIGS) are studied within a single laboratory. This arrangement offers the potential to unlock significant synergies in many topics common to all thin-film-based technologies.

PVcomB – as part of the Helmholtz-Zentrum Berlin (HZB) and in close cooperation with the Technical University Berlin (TUB), the University of Applied Sciences Berlin (HTW) and industrial partners – combines competences in fundamental materials research and device development on the one hand, with industrial experience and technology on the other.

Work at PVcomB concentrates not only on upscaling of promising device concepts from fundamental research, but also on existing industrial processes. For this, PVcomB has created a semi-industrial-like environment to also address aspects of throughput, statistics, reliability and 'easy-to-transfer processes.' The two production 'baselines' for TF PV modules offer the ideal references for new materials, process steps and methods: new device concepts can be directly compared to state-of-the-art technology. In the same way, technological questions from industry (e.g. producers of solar modules as well as manufacturers of production equipment) can be handled by implementing alternative materials, layers or process steps and compared to the reference process.

The two reference lines – PVcomB's backbone

In 2009 PVcomB was recognized as the leading PV cluster in Germany. Over a five-year period (2010–2014), PVcomB will receive €15 million in funding from the BMBF (German Federal Ministry of Education and Research) and the state of Berlin. An advanced research infrastructure has been set up with this support.

Fig. 1 shows a schematic of the two baselines. The heart of the TF Si line is a plasma-enhanced chemical vapour deposition (PECVD) cluster tool – the AKT 1600 from Applied Materials. This tool is equipped with three deposition chambers and a vacuum magazine for up to six 30×30cm^2 glass substrates. A throughput of up to ten tandem stacks per day is achieved from 12 hours' average operation. An in-line sputter tool (A600V7) from Leybold Optics Dresden is used to develop transparent conducting oxide (TCO) front contacts and to prepare TCO/metal back contacts.

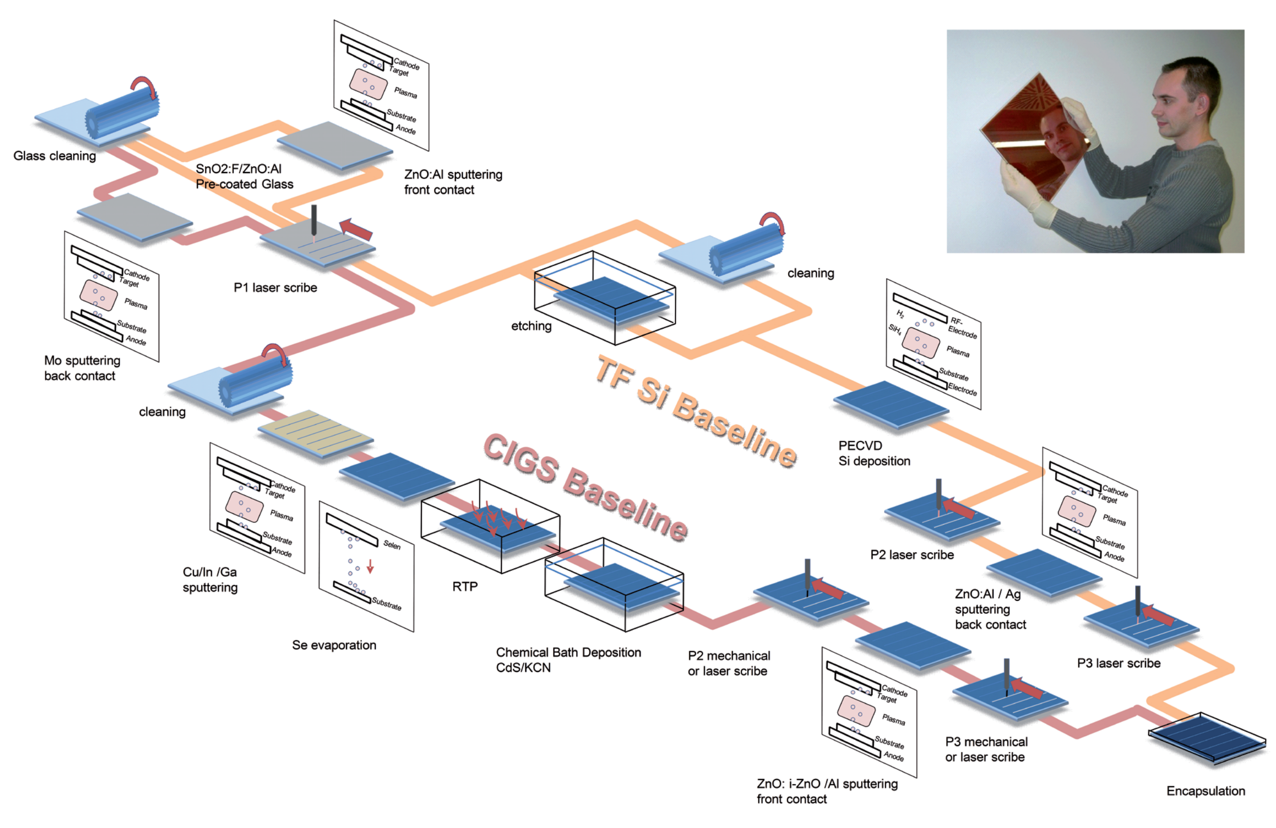

Figure 1. Schematic representation of the two 30×30cm² reference lines for TF Si (a-Si/µc-Si) and CIGS modules.

Four magnetron positions (two planar, two rotatable) can be used for various materials. A sixfold carrier magazine for changing substrates and having the ability to coat two substrates simultaneously ensures that automated processes can be run with a high throughput and a high level of reproducibility.

A high-performance laser-scribing tool from Rofin Baasel Lasertech is used to prepare baseline laser scribing as well as for developing novel concepts of laser patterning. Its configuration with nano- and picosecond lasers is optimized for both applications.

The CIGS baseline follows the sequential processing concept. A second A600V7 in-line sputtering tool (four planar and two rotatable magnetrons) is used to deposit the back contact (Mo, SiO_x) and the precursor layers of the absorber (CuGa, In). After a subsequent deposition of selenium, the layer stack is annealed in a rapid thermal processing furnace from Centrotherm. Advanced chemical labs are set up to perform the automatic chemical processing (chemical bath deposition and etching) before the TCO front contact is deposited in a VIS300 sputtering tool from Von Ardenne Anlagentechnik. The above-mentioned Rofin laser tool is also used for all scribes within the CIGS line. It is also equipped with a mechanical scribing tool in order to be comparable to the industrial standards for patterning.

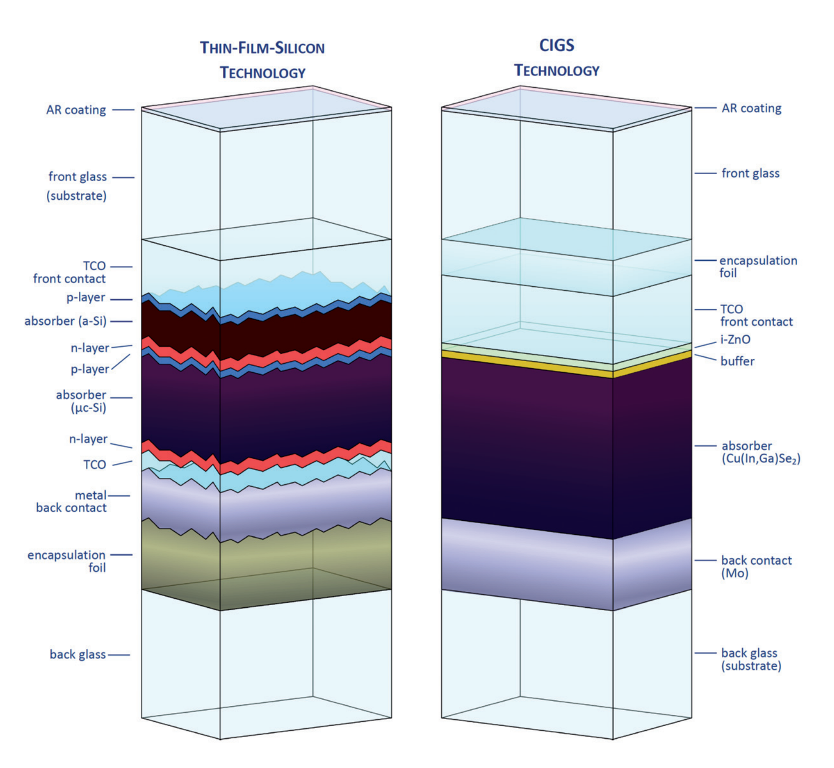

Figure 2. Layer structure of the solar cells developed at PVcomB: a-Si/µc-Si tandem cell (left) and CIGS (right). The entire layer stack, excluding glass and encapsulation, is typically 2–3µm thick.

To optimize preparation processes and device behaviour, PVcomB has set up a comprehensive data management system. Real-time investigations of processes (e.g. plasma analysis, residual gas analysis) are combined with tool-side process parameters (e.g. temperatures, gas flows): this allows short feedback times and an excellent understanding of deposition processes. Process tools as well as test sites are attached to an electronic lab book system; with this, operators and

researchers have fast access to all relevant parameters and results. To ensure a strictly confidential workflow, all selected data are protected by a reliable access control system.

Thin-film silicon solar cells and modules

Thin-film silicon solar cells are dominated by amorphous silicon (a-Si) p-i-n single junctions and amorphous/microcrystalline silicon (a-Si/μc-Si) p-i-n/p-i-n tandem junctions (Micromorph). With a tandem junction (see Fig. 2), an a-Si top cell of thickness 0.2–0.3μm is followed by a μc-Si bottom cell of thickness 1–2μm. While the top cell transforms the visual (VIS) fraction of the sun spectrum at a high voltage of ~900mV, the bottom cell transforms the longer wavelength fraction (near-infrared, NIR) at ~500mV. Because of light-induced defect generation in a-Si, the initial conversion efficiency η of tandem cells drops by typically 10–12% (relative) upon light-soaking before it stabilizes. For lab cells, η_{stable} = 12.3% for a tandem junction and η_{stable} = 10.1% for a single junction have been demonstrated by Oerlikon [1,2]. Owing to the higher conversion efficiency compared to a-Si single junctions, the tandem junction is becoming increasingly dominant in the thin-film silicon PV industry. Large-scale tandem modules achieve η_{stable} of 10–11% [3,4]. One of the main challenges in increasing η further is the limited light absorption in the thin μc-Si layer because of the low absorption coefficient of crystalline silicon. Sophisticated light-trapping schemes and textured substrates are therefore being developed. Here, the correct property of the TCO is crucial. At PVcomB, tandem cell processes have been established for all commonly used TCO substrates (SnO_2:F, ZnO:Al, ZnO:B). New transparent materials and better light-scattering substrates and back contacts are currently being developed.

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Fig. 3 shows the results obtained at PVcomB for the initial efficiency over time of a-Si/μc-Si cells deposited on two different large-scale TCO substrates: commercial SnO_2:F (FTO) (blue) and DC magnetron sputtered and texture-etched ZnO:Al (AZO) (red). Both TCOs are on low-iron

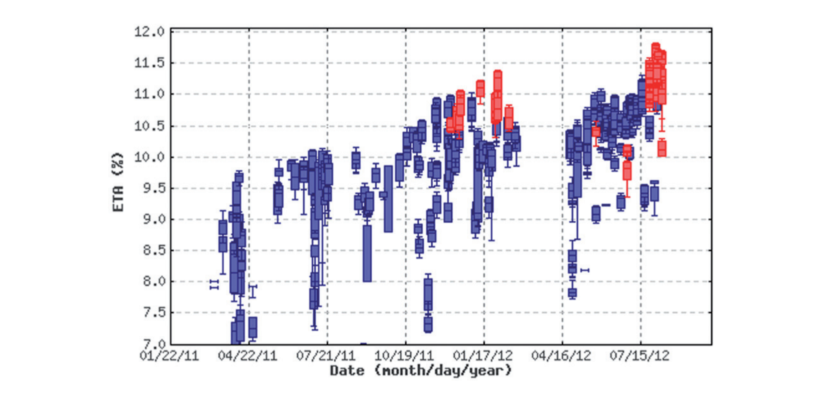

Figure 3. Box plot with a timeline of a-Si/μc-Si (initial) tandem cell efficiency values realized at PVcomB for different TCO substrates: FTO (blue) and AZO (red). A two-month downtime for maintenance and hardware modifications can be seen in spring 2012.

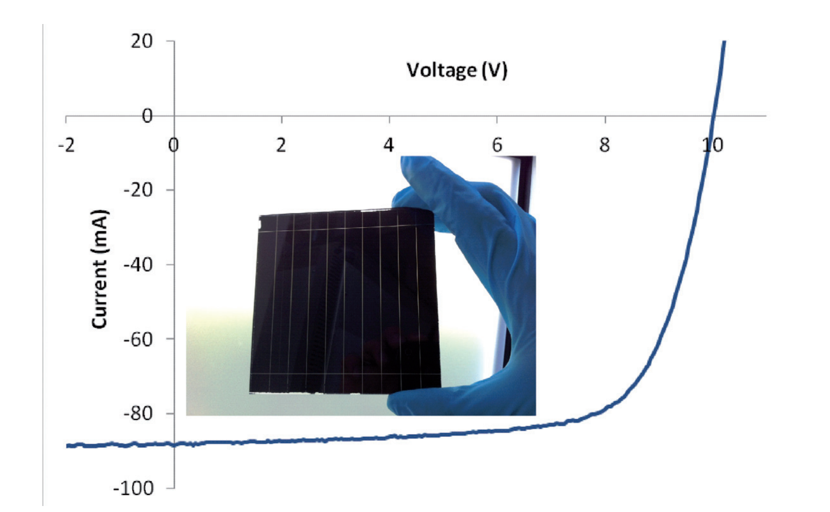

Figure 4. *I-V* characteristics of an a-Si/μc-Si mini-module (seven cells, 56cm² aperture area) from the PVcomB baseline, with 11.3% (initial) efficiency.

3.2mm-thick float glass without anti-reflection (AR) coating. During the last year of operation, the efficiency was increased by more than 2% absolute. So far, the best cell has achieved 12% (initial) and 10.8% (after 168 hours of light-soaking at AM1.5, 50°C), both on ZnO:Al. For recent 10×10cm² mini-modules (180μm interconnection width dead area), an initial efficiency of 11.3% was achieved (Fig. 4) – this is expected to stabilize at well over 10%.

Much higher efficiencies, however, are required. Most research groups currently working on a-Si/μc-Si tandems are therefore focusing on three topics:

1. Highly transparent (and reflecting) doped p- and n-layers, e.g. based on μc-SiO_x.
2. Low-defect growth of μc-Si:H on highly textured substrates.
3. Highly transparent, multi-scale, textured TCO substrates.

These three topics are being addressed at PVcomB, and the improved processes have been developed to be implemented in our baseline, demonstrating robustness, scalability and compatibility with industrial production. The latter is done in cooperation with our industry partners. It is aimed to demonstrate mini-modules with a stable efficiency of 12%. Examples of this work are given next.

New doped layers

In the most recent tandem cell shown in Fig. 2, all doped layers are based on μc-SiO_x, a mixed-phase material consisting of a (doped) μc-Si phase embedded in a-SiO_x as matrix material. Originally proposed and implemented by Kaneka Corp., it has been developed for cells over the last few years [5,6]. Besides the reduced parasitic absorption as compared to a-Si or μc-Si doped layers, the refractive index n can be varied in a wide range, which allows μc-SiO_x layers with low $n \approx 1.7$–2 to be employed as reflective layers (at the internal n/p contact)), or, with high $n \approx 2.5$, as an anti-reflective front p layer. Furthermore, the negative influence of local micro shunts on the cell's *FF* and V_{oc} is believed to be suppressed as a result of limited lateral conductivity.

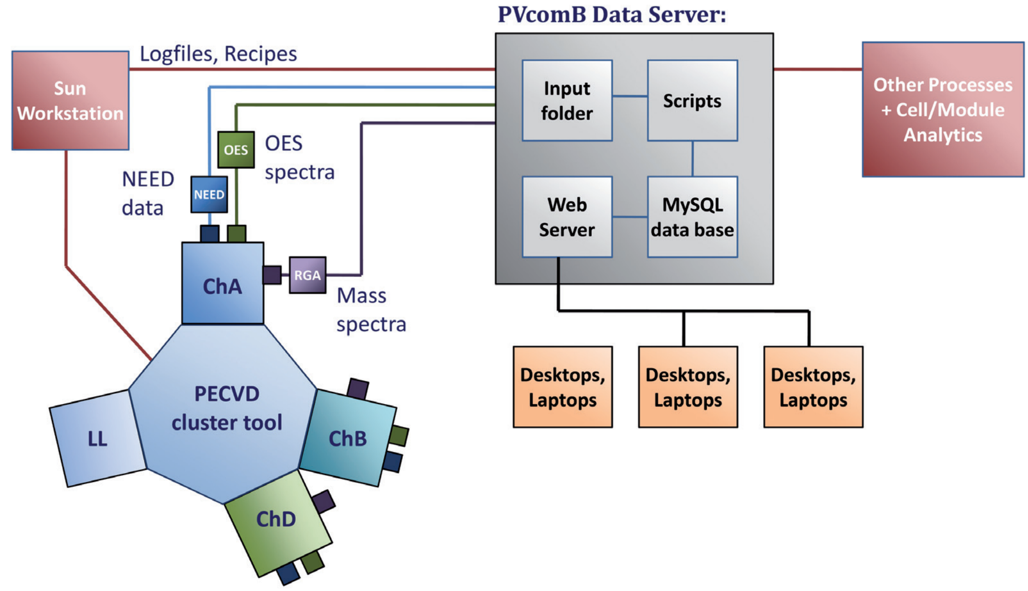

Figure 5. Schematic of the PECVD cluster tool (AKT), with in situ diagnostics and connection to the in-house database system.

The focus is on further development of that material in addition to establishing and transferring a robust process to large-scale production in collaboration with our industrial partners (Masdar PV and Inventux). Moreover, within the Helmholtz-Zentrum Berlin, research is being carried out on new materials based on SiO_x, SiC_x and SiN_x to be used as functional layers, for example doped layers, or as barrier layers for high-T applications, such as polycrystalline Si cells [7,8].

Highly transparent TCO

As can be clearly seen in Fig. 3, the cells based on ZnO:Al TCO outperform the cells on SnO_2:F, which partially results from better AZO transparency and light scattering, and partially from an optimized and more transparent AZO/p design based on the above-mentioned μc-SiO_x. A method for significantly improving the properties of ZnO:Al by annealing the TCO at 500–600°C under a thin a-Si capping [9,10] has been developed at Helmholtz-Zentrum Berlin. The free-carrier mobility was thus increased from about 35 to more than 70cm²/Vs, helping to reduce absorption in the NIR. Moreover, transmission in the VIS is improved as a result of the annealing of structural defects at the grain boundaries. It was recently demonstrated at PVcomB that this leads to an improved short-circuit current. The quantum efficiency of initial cells on annealed ZnO:Al has already shown a 5% increase in current compared to the best cells on standard ZnO:Al TCO [11].

PECVD in situ diagnostics & in-house database

As mentioned above, PVcomB operates a PECVD cluster tool that is specifically designed for industrial applications; high throughput and reliability are key properties of this tool. This is crucial in order to optimize and control the baseline and to run the various R&D projects with enough statistics on one tool. To gain an understanding of growth processes, such as μc-Si or μc-SiO_x, as well as NF_3 etching and chamber conditioning, PECVD in situ diagnostics were implemented: OES (optical emission spectroscopy), RGA (residual gas analyzer, mass spectrometry) and a Hercules sensor by Plasmetrex for measuring the electron dynamics (NEED) of the plasma. These three techniques are complementary to, as well as compatible with, industrial production. With a database system developed in-house, all PECVD recipes and deposition parameters, in addition to the in situ diagnostics data, are logged, with easy access via a user interface (shown schematically in Fig. 5) [12]. Moreover, the database contains *I-V* and EQE (external quantum efficiency) measurement results from cells and modules. This facilitates the easy correlation of process data and plasma properties with cell parameters. Data mining and statistical process control, as well as fast troubleshooting, is also made possible.

CIGS solar cells and modules

Cu(In,Ga)$(S,Se)_2$ (CIGS) solar cells hold the technology world record in efficiency in the field of thin-film photovoltaics, with cell efficiencies of up to 20.3% [13]. There are several approaches to manufacturing CIGS solar modules: they can be classified as *sequential processing* and *co-evaporation*. In the case of co-evaporation, all elements are evaporated in a single vacuum chamber to form the CIGS layer; sequential processing, on the other hand, begins with a Cu/In/Ga precursor layer, which is converted in an annealing step into CIGS. Nevertheless, for both of these technologies, it has not yet been possible to transfer the record efficiencies into mass production, as the homogeneity in terms of composition and thickness of the CIGS layer plays a crucial role.

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As of July 2012, all baseline equipment for CIGS solar cell processing has now been delivered and production has started. Results from the first solar cells are presented in the following section. As described earlier, the CIGS baseline applies a typical sequential processing sequence [14]. A substrate soda-lime glass is coated with an 800nm single-layer molybdenum (Mo) back-contact layer. Subsequently, a stacked elemental layer (SEL) of 370nm Cu:Ga, 500nm In (by magnetron sputtering) and Se (by thermal evaporation in a vacuum chamber) is deposited. This SEL is converted into the CIGS absorber layer within 90 seconds in a rapid thermal processing (RTP) furnace with radiation heating at 570°C. The temperature ramp is about 4K/s within the CFC carrier box. The whole process time is approximately 4 minutes (without pump- and cool-down times). After the p-type CIGS absorber layer is formed, the

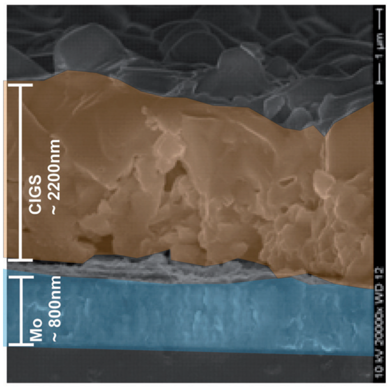

Figure 6. SEM cross section of a CIGS absorber layer.

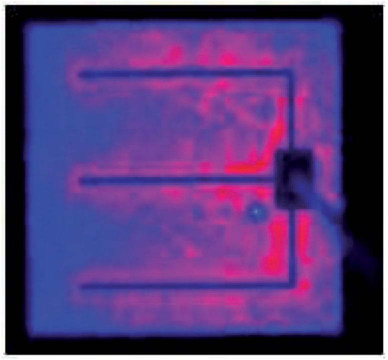

Figure 7. IR thermography image of a CIGS solar cell.

n-type CdS buffer layer is deposited in a chemical bath deposition (CBD). The cell is finished by the application of an intrinsic ZnO (i-ZnO)/ZnO:Al double layer. To produce a simple sample preparation, the 30×30cm^2 substrates are first cut into nine 10×10cm^2 coupons. These coupons are divided into 49 cells of 1.4cm^2. The cells are then contacted via a Ni/Al/Ni contact grid deposited by electron-beam evaporation.

The initial layers are thoroughly analyzed with respect to their compositional and thickness homogeneity, morphology and solar cell parameters. Absorber composition and thickness of precursor and absorber layers are determined by X-ray fluorescence spectroscopy (XRF), measured by a Rigaku XRF Primus II instrument, using a first-principles method without a calibration sample. The homogeneity is calculated by taking nine measurement points equally distributed on the 30×30cm^2 substrate. The thickness inhomogeneity of the sputtered precursor layers is about 2% for Mo and Cu:Ga, and 3.6% for In. Selenium is deposited with a thickness variation of about 10%. The resulting absorber layers have a Cu/(In+Ga) ratio of 0.82, with a variation of only 5.1%, while the absorber thickness varies between 1.94µm at the edge and 2.25µm in the centre (roughly 12% variation).

The best solar cell from our first run yielded a conversion efficiency of 8.7%, with a short-circuit current density (J_{sc}) of 40.2mA/cm^2, an open-circuit voltage (V_{oc}) of 400mV and a fill factor (*FF*) of 54%. The low fill factor can be explained by a high series resistance (R_{oc}) of 4Ωcm^2, which is mainly caused by the use of a ZnO window layer that was optimized for another CIGS device, not for the PVcomB stack. The low V_{oc} indicates that no gallium is at the top of the absorber layer, as gallium increases the band gap of CIGS and therefore the open-circuit voltage.

Scanning electron microscope (SEM) cross-section imaging reveals a compact and dense layer with no large holes (Fig. 6). Nevertheless, the polycrystalline CIGS has larger grains at the top and smaller grains at the bottom. Energy dispersive X-ray spectroscopy (EDX) line scans detect a large quantity of Ga in the small grains at the Mo/CIGS interface, indicating that these are Ga-rich crystallites. The agglomerate of gallium at the Mo back contact is typical for a sequential CIGS process. Pushing the Ga to the top is the biggest challenge in future process development. Quantum efficiency (QE) measurements also indicate that the top layer is pure $CuInSe_2$, since QE is still above 50% at 1200nm, which correlates to a band gap of 1eV, as expected for $CuInSe_2$. This is also the reason why the best initial solar cells have a V_{oc} of only 400mV.

Infrared (IR) lock-in thermography imaging shows only a few shunts in the solar cells (Fig. 7). This supports the image of a dense and homogeneous CIGS layer structure produced in the first experiments within the CIGS baseline.

In parallel with the optimization of CIGS solar cell processing, PVcomB is also targeting new concepts to improve the solar cell quality. Three major topics will be investigated:

1. A barrier layer to prevent uncontrolled sodium diffusion from the glass substrate, plus the application of additional sodium diffusion, such as a Mo back contact doped with sodium.
2. A Cd-free buffer layer using dry processes, such as atomic layer deposition (ALD).
3. The development of elemental selenium and sulphur sources for the RTP process in order to achieve a better control of the CIGS growth process.

The joint laser laboratory of HTW and PVcomB

In thin-film solar cell manufacturing, further reduction in cost, as well as increase in yield efficiency, requires the most innovative concepts. The application of laser technologies is already well established in the industrial production process (for example for monolithic series interconnection) and helps to drive down the cost per W_p. The three scribing steps P1, P2 and P3, alternating with thin-film deposition, are necessary for creating a serial interconnection of many cells on a single substrate.

For further increases in module efficiency and production yield, high precision patterning is required. To meet these challenges, HTW (with its very strong laser background) and HZB have joined forces in PVcomB to set up a state-of-the-art laser processing system and to develop new solar cell concepts.

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The laser-based patterning tool of HTW, directly integrated in PVcomB's laboratory environment, is equipped with a high-speed motion system and two different

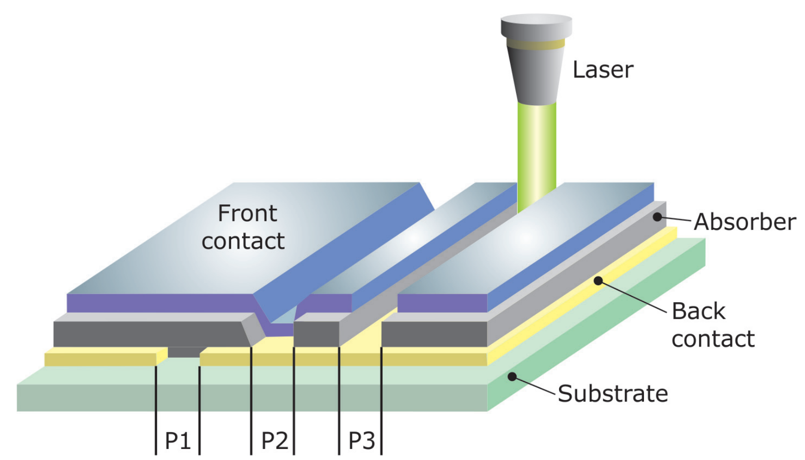

Figure 8. Schematic of a typical monolithic series interconnection of TF solar cells.

laser sources. The motion system facilitates accurate patterning with deviations of less than 5µm at velocities of up to 1200mm/s. This high accuracy allows us to reduce the area losses resulting from serial interconnection. Well-established standard patterning is performed using a nanosecond laser source with a wavelength of 532nm. For the purposes of developing novel concepts as well as heat-sensitive patterning, a picosecond laser source with a wavelength of 1064nm, in addition to second- and third-harmonic generation of laser pulses with 532nm and 355nm wavelengths, is integrated. For reference purposes, a scribing needle is available.

This innovative set-up allows novel laser-based patterning concepts to be developed and established for industrial solar cell fabrication as well as for the upscaling of these processes. It is also possible for solutions to be developed for external partners for their industrial laser applications, such as wafer marking, or for evaluating suitable parameters for patterning these materials.

At present, research topics include complete laser patterning of CIGS cells, especially patterning with a single nanosecond wavelength [15], and simulation of the laser–matter interaction in stacked layer systems [16]. Further investigations to seek a fundamental understanding of laser–matter interaction, such as the molybdenum ablation behaviour, are in progress.

Advanced analytics for device and process optimization

The analytics baseline at PVcomB comprises a number of standard characterization methods that are applied to, and documented online for, each processed sample. The equipment is calibrated on a regular basis to ensure highly reliable results in conformance with international standards. The backbone of the standard methods is formed of the AAA dual-source WACOM Solar Simulator and the AAA dual-source h.a.l.m. Flasher. These are complemented by a dual-source EQE set-up and a detailed optical characterization by means of a Perkin Elmer high-performance Lambda 1050 spectrophotometer. Apart from the standard tests, the continuous further development of analytical equipment, along with the implementation of new tools, is of high importance. Currently the integration of a lock-in thermography system (by Thermosensorik) and an electroluminescence set-up (by Great Eyes) in the baseline analytics process is the centre of attention (see Fig. 7). In addition, a number of methods – such as angle-resolved scattering (ARS), Raman spectroscopy, Hall effect and illuminated lock-in thermography – are available on site for basic research of materials.

With regard to advanced thin-film photovoltaic device analytics, a close cooperation has been established between the initial PVcomB partners and the Joint Lab (JL) of IHP Frankfurt/O and BTU Cottbus. As a result, a wide range of sophisticated analytical techniques suitable for PV materials is accessible for PVcomB research.

The PVcomB research at JL focuses on the adaptation and application of transmission electron microscopy (TEM) methods, in addition to spatially and spectrally resolved photoluminescence (PL) [17] techniques, each of which requires an approach to compete with the side effects of the glass substrate and low signal intensity. These methods are complemented by the Hamamatsu PHEMOS 1000 set-up at TU Berlin for spatially and spectrally resolved electroluminescence (EL) as well as light-beam-induced current (LBIC) investigations. In particular the latter can give valuable information on light-scattering effects in thin-film photovoltaic devices [18].

Among the methods available at TU Berlin, X-ray fluorescence techniques – such as grazing incidence XRF (GIXRF) – have already been successfully applied to both chalcopyrite and silicon absorber systems [19,20]. Here, a non-destructive depth-resolved analysis of the chemical composition is obtained by varying the X-ray angle of incidence, thus making it possible to extract information about, for example, composition gradients or buried interface properties. As mentioned earlier, PVcomB is equipped on site with a Rigaku WD-XRF ZSX Primus II. Because of the wavelength dispersive detection set-up, elements down to boron (Z=5) can be detected with high sensitivity and reproducibility. The implemented layer model is well adapted to a fundamental parameter-based analysis of the typical precursor stacks in chalcopyrite technology, yielding the thickness as well as the composition of individual layers. Last, but not least, the Primus II system comes with a mapping facility that provides information on lateral compositional non-uniformities.

In order to understand and distinguish the manifold effects caused by the thin-film nature of thin-film photovoltaics, PVcomB has finally entered the field of 2D/3D device modelling. There are several suitable commercial and non-commercial simulation tools available on the market, each offering a wide variety of physical models and pre-selected parameter sets (usually in regard to crystalline silicon). However, in order to obtain reliable information, the relevant models and parameters have to be chosen very carefully. PVcomB's approach is to use the knowledge gained by years of 1D modelling for a step-wise expansion of the 1D solar cell model into space. This extension is essential for a proper description of the complex optoelectronic interaction in current thin-film devices. Light-management phenomena [21] and functional non-uniformities, for example in intermediate reflector concepts, accompany advanced contacting schemes that can be effectively optimized by device modelling [22].

> "The mission is to bridge the gap between innovative lab-sized solar cell concepts and industrially produced modules."

Summary

PVcomB is a joint initiative of HZB and the Technical University Berlin. Together with HTW and other partners from research and industry, a unique institution has been set up for applied research and development in the field of thin-film PV. The mission is to bridge the gap between innovative lab-sized solar cell concepts and industrially produced modules. To this end, PVcomB has set up two dedicated reference lines for research in thin-film Si- and CIGS-based modules with an area of 30×30cm^2; this module size is well suited to addressing various issues arising in industrial production. State-of-the-art TF modules from both of these technological routes are developed and produced in a semi-industrial environment, for investigating issues such as process stability, throughput, statistics and reliability. Advanced analytical methods for in situ and ex situ characterization of materials and devices have been established to support the whole range, from baseline characterization to advanced fundamental research. An ideal reference has therefore been created for the implementation of new materials, process steps and technologies.

Acknowledgements

We would like to acknowledge the invaluable contribution of all persons involved in building up PVcomB, from the initial idea generation to establishing full functionality. We also wish to express our appreciation for the support from our partner companies and institutions. This work was supported by the Federal Ministry of Education and Research (BMBF) and the state government of Berlin (SENBWF) in the framework of the 'Spitzenforschung und Innovation in den Neuen Ländern' programme (grant no. 03IS2151).

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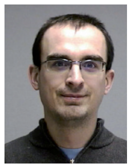

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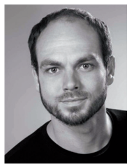

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