MICRO CONCENTRATOR CONCEPT FOR COST REDUCTION AND EFFICIENCY ENHANCEMENT OF THIN-FILM CHALCOPYRITE PHOTOVOLTAICS: RESULTS FROM EU JOINT RESEARCH PROGRAM CHEETAH

M. Schmid[1], E. Lotter[2], X. Lin[1], L. Wang[1], R. Klenk[1], K. Eylers[3], F. Ringleb[3], T. Boeck[3], B. Heidmann[1], G. Nenna[4], F. Loffredo[4], F. Villani[4], T. Köhler[1], D. Sancho-Martinez[1], T. Raadik[5], J. Krustok[5], M. Grossberg[5], M.Ch. Lux-Steiner[1]
[1]Helmholtz-Zentrum Berlin für Materialien und Energie, Hahn-Meitner-Platz 1, 14109 Berlin, Germany
[2]Zentrum für Sonnenenergie- und Wasserstoff-Forschung Baden-Württemberg, Meitnerstr. 1, 70563 Stuttgart, Germany
[3]Leibniz-Institute for Crystal Growth, Max-Born-Straße 2, 12489 Berlin, Germany
[4] ENEA Portici Research Center, P.le Enrico Fermi, 80055 Portici (Naples), Italy
[5]Tallinn University of Technology, Ehitajate tee 5, 19086 Tallinn, Estonia

ABSTRACT: Results for research on chalcopyrite micro concentrator solar cells obtained within the framework of CHEETAH joint program are shown. A top-down proof of concept study reveals close to 30% relative efficiency increase under light concentration using inkjet-printed CIGSSe. A novel bottom-up approach for local chalcopyrite absorbers grown from indium islands demonstrates working CISe micro cells. Millimeter-sized lenses are fabricated from PMMA by a casting process to be applied as concentrator optics. For a combined exploitation of direct and diffuse light components an angular splitting concentrator based on chalcopyrite and kesterite absorber material is proposed. The scientific innovation brought will enrich further development of CIGSe solar cells and contribute to their relevance in photovoltaic energy production.

Keywords: Concentrator Cells, Design, Fabrication, Chalcopyrite, Cu(InGa)Se_2, Kesterite, CZTS, Lenses

1 BACKGROUND

The European SET-Plan envisages an accelerated development of low-carbon technologies, including photovoltaics (PV). The related aim of reduced material consumption and enhanced efficiency followed in current PV research is reflected in the goals of the joint research program “Cost-reduction through material optimisation and Higher EnErgy output of solAr pHotovoltaic modules - joining Europe’s Research and Development efforts in support of its PV industry” (CHEETAH). Within work package 9, task 2 focuses on thin-film chalcopyrite (Cu(In,Ga)Se_2 – CIGSe) solar cells and addresses their challenge of containing indium and gallium - elements with high supply risk, which hinders CIGSe to enter production on the terawatt scale. The innovative approach to face this challenge and fabricate CIGSe solar cells with higher efficiency and less material is the application of a micro concentrator concept. By restricting the solar cell dimensions to a few ten to hundred microns in diameter and reducing the coverage with absorber material by a factor of about one hundred, tremendous material saving is possible. Simultaneous concentration of incident sun light onto the reduced absorber areas allows to fully exploit incident radiation and benefit from enhanced performance due to increased illumination energy density. The dimensionality on the microscale enables improved heat dissipation compared to large scale counterparts and opens the path towards fabrication of solar concentrator modules with a compactness comparable to regular thin film products.

In this contribution we show results of CHEETAH joint research on chalcopyrite micro concentrator solar cells: In a top-down proof of concept study, close to 30% relative efficiency increase is achieved by light concentration onto inkjet-printed CIGSSe. Using a novel bottom-up approach for local chalcopyrite growth from indium islands we demonstrate working CISe micro cells. Corresponding millimeter-sized lenses are fabricated from PMMA by a casting process. Finally, we propose an angular splitting concentrator exploiting both direct and diffuse light components by combining chalcopyrite and kesterite absorber material. The related scientific innovation and progress will strengthen further development of CIGSe solar cells and contribute to make them a vital part of photovoltaic energy production.

2 RESULTS AND DISCUSSION

2.1 Top-down micro cells based on inkjet-printed CIGSSe

Material deposition by inkjet-printing offers an ultimate approach for full spatial control over absorber growth. The formation of planar CIGSSe absorbers by inkjet-printing leading to a solar cell efficiency of 11.3% has been reported previously [1]. In brief, a precursor ink containing Cu, In and Ga is formulated, printed by drop-on-demand technology onto a Mo back contact and

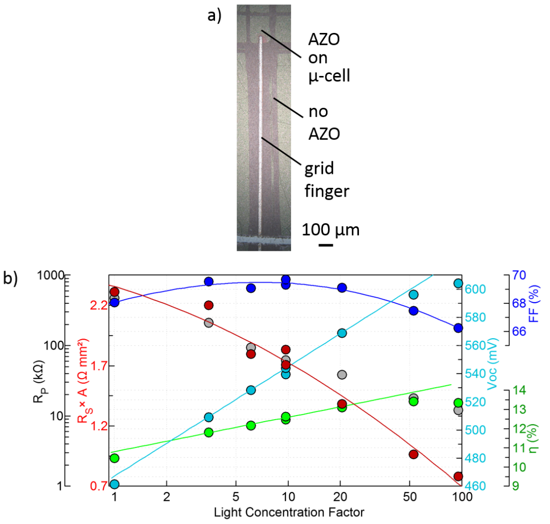

Figure 1: a) Micro solar cell defined on planar inkjet-printed CIGSSe absorber by local etching of ZnO:Al (AZO) front contact, b) corresponding electrical solar cell parameters as a function of light concentration.

subject to a three-stage chalcogenization process in selenium-containing atmosphere and with addition of H_2S. Whilst on the long run a bottom-up approach by local absorber printing is feasible, for the proof of concept study the micro cells were fabricated by the following top-down approach based on planar CIGSSe solar cells: the front part of a Ni-Al grid finger was mechanically separated and a micro cell defined around the tip by chemical etching of the surrounding ZnO:Al front contact, which was also removed along the grid finger. The resulting structure is depicted in Fig. 1a). Fig. 1b) shows the performance of an according micro cell under homogeneously enhanced light illumination. The measured short-circuit current density is assumed to increase linearly with concentration and is used to define the light concentration factor. Up to about 60 times concentration the open-circuit voltage V_{OC} follows the expected logarithmic increase and improves from 460 to 600 mV. Combined with small variations in fill factor *FF* only, the efficiency reaches its maximum at 60 suns as well: $\eta = 13.4\%$ efficiency is obtained which corresponds to a relative increase of 28% compared to $\eta = 10.5\%$ at one sun. Remarkably, the development of series and shunt resistance, R_p and R_s, respectively, reveals a different behavior for different absorber materials – compare the results for micro cells fabricated from co-evaporated CIGSe shown in [2] – and will require further analysis to obtain highly efficient micro concentrator solar cells.

2.2 Bottom-up growth of CISe micro cells from In droplets

True material saving can only be achieved if a bottom-up approach is followed for the micro solar cell fabrication. Addressing in particular a reduced consumption of the rare and expensive element indium, we locally deposit In droplets, which are further processed to CISe absorbers. The growth and regular arrangement of In droplets on laser-patterned substrates has in detail been described elsewhere [3]. Droplets tuned to a diameter of about 40 µm and a thickness of few microns are then overcoated with a 500 nm thick Cu layer. A subsequent selenization process in ramps of 200°C and 550°C leads to the formation of CISe and $CuSe_x$, the latter being removable by selective etching. Fig. 2a) shows an SEM top view of a resulting CISe micro absorber island. In Fig. 2b) the corresponding structure of the micro solar device is represented. Individual absorber islands grown on molybdenum back contact were electrically isolated by SU-8 photoresist, which was deposited by spin-coating and removed on top of the CISe islands by plasma etching. In this way the CdS, i-ZnO ad ZnO:Al front contact layers applied to the entire surface are forming the pn-junction with the local CISe absorber without touching the Mo back contact, present also in between the absorber islands. Resulting dark and illuminated current-voltage characteristics clearly revealing a diode behavior are shown in Fig. 2c). Even though there is still room for improvement regarding the solar cell's performance parameters, the achievement of a working locally grown chalcopyrite solar cell is an important step towards material saving and efficiency enhancement.

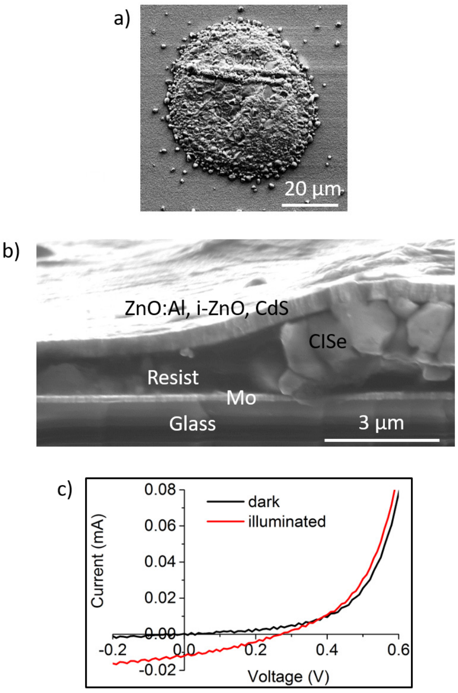

Figure 2: CISe micro solar cell fabricated by a bottom-up process: a) SEM top view of CISe absorber, b) cross sectional SEM image of isolated micro solar cell, c) dark and illuminated current-voltage characteristic.

2.3 Inkjet-printed PMMA micro lenses

Asides the formation of local CIGSe absorbers the fabrication of adequate lenses is essential to obtain micro concentrator devices. Inkjet-printing mentioned above for

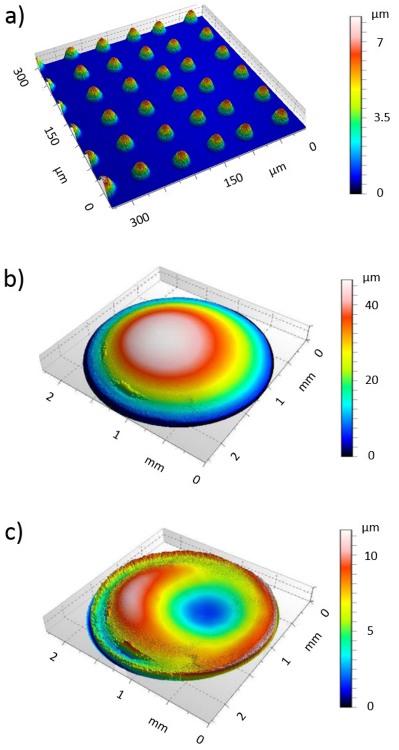

Figure 3: PMMA lenses: a) micrometer-scale array, b) millimeter-scale single optical element revealing deviation from spherical shape as visible in c) corresponding error profile error.

the absorber growth can also be used for the deposition of concentrator optics, enabling on the long run an all-printed device. State-of-the-art inkjet-printed lenses made from poly(methyl methacrylate) (PMMA) usually reach diameters on the order of several tens of microns. An example of a micro lens array deposited by drop-on-demand inkjet printing technology of a high concentration PMMA-based ink onto a 325 x 325 µm^2 substrate is depicted in Fig. 3a). The substrate temperature was kept at 17°C during the printing process and for the subsequent 30 minutes; then the samples were annealed at different temperatures. The results suggest that 250°C is the optimized annealing temperature allowing to reduce the surface roughness and to improve the spherical shape. These findings are significant to produce high-quality optical microstructures. Slightly bigger optical structures than those displayed in Fig. 3a) and having dimensions up to hundreds of microns were manufactured through non-conventional printing technologies (pyro-electrohydrodynamic inkjet printing) by dispensing high-viscosity optical-grade polymer based inks [4]. Having to couple the micro lenses with micro solar cells of approximately 100 µm diameter and reaching concentration factors of 100 -500 times, lens dimensions in the range from 1 - 2.3 mm are needed. Such large dimensions have never been reported for inkjet-printed PMMA lenses in literature due to process constraints. Therefore, micro lenses were manufactured by a casting process via a chemical pipette allowing to deposit the required ink volume in a single droplet. An example of the so-manufactured optical elements having a diameter of 2.2 mm and a maximum height of 46 µm is shown in Fig. 3b). A remaining deviation from the perfect spherical shape, as illustrated by the error profile displayed in Fig. 3c), needs to be addressed in further studies for final application to micro solar cells making integrated concentrator devices feasible.

2.4 CIGSe-CZTS direct-diffuse micro concentrator

As an alternative absorber for micro concentrator solar cells, kesterite Cu_2ZnSnS_4 (CZTS) material was taken into consideration. Testing of CZTS monograin layer solar cells [5] under increased illumination intensities, however, yielded a decrease rather than an increase in efficiency. Fig. 4a) reveals the major reason, namely a significant drop in fill factor (*FF*) with enhanced illumination intensity above one sun, which directly translates to a decrease in efficiency (not light soaked, room-temperature measurements). Therefore, CZTS solar cells are not suitable for operation under concentrated sun light. In contrast, a reduced illumination intensity improves their performance as shown in Fig. 4b). When decreasing the light intensity from 1 to 0.15 suns the *FF* increases by 11% absolute, which finally leads to an increase in efficiency from 4.0% to 4.6% in the example shown here. Kesterite material thus appears predestined to be operated under reduced rather than enhanced illumination intensity. This observation lead to the development of a combined CIGSe-CZTS direct-diffuse micro concentrator concept. The concept takes into account that concentrator optics are only able to focus direct irradiation, whereas the significant part of diffuse light contained in the solar radiation incident in our latitudes cannot be concentrated. Exploitation of both radial parts of the solar spectrum is aspired by placing CIGSe micro cells in the concentration regions and filling the spaces in between the micro cells by CZTS cells such that best performance enhancement of both types of solar cells under increased and reduced illumination intensity, respectively, is achieved. The improved efficiency of CIGSe solar cells with increased illumination intensity was already proven in sec. 2.1; for the striped CIGSe micro cells of the angular splitting concentrator planar co-evaporated absorbers were locally passivation and shaded. Fig. 4c) shows the schematic structure of the combined device and Fig. 4d) the prototype, which could confirm the expected performance behavior.

3 SUMMARY

As the results presented here reveal, significant progress has been achieved with respect to top-down and bottom-up formation of micro absorbers from planar inkjet-printed CIGSSe and sequentially processed CISe based on indium islands, respectively. Furthermore, the development of printed polymer lenses on the millimeter-scale was reported and a novel concept for enhanced exploitation of direct and diffuse sun light proposed. The related joint work within the European project CHEETAH therefore constitutes an important resource of combined forces to advance the development of highly efficient and low cost photovoltaics.

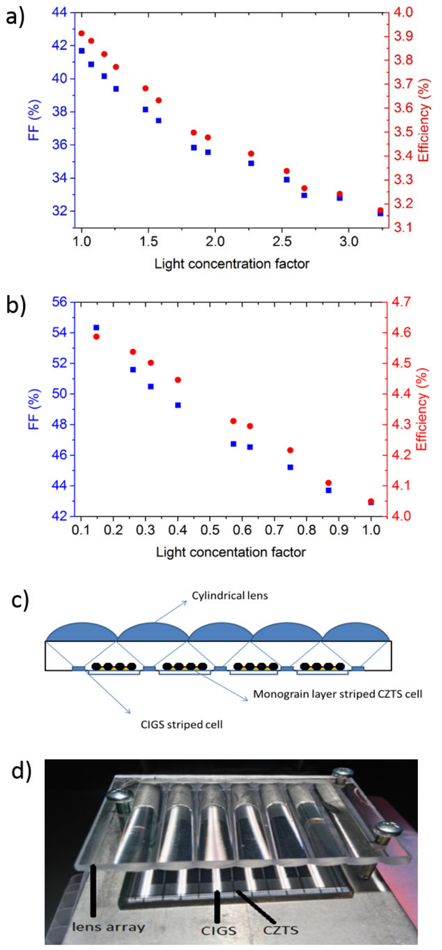

Figure 4: FF and efficiency revealing CZTS solar cell performance under a) enhanced and b) reduced illumination conditions; c) schematic and d) prototype of combined CIGSe-CZTS direct-diffuse micro concentrator concept.

ACKNOWLEDGMENT

This research is part of the EU CHEETAH project and has received funding from the European Union’s Seventh Programme for research, technological development and demonstration under grant agreement No 609788.

The authors would like to thank M. Kirsch (HZB) for ZnO sputtering and grid deposition onto CIGSe cells as well as O. Ernst (IKZ) for preparation of resist-based isolating layers. We thank R. Miscioscia (ENEA) for optical profilometer measurements and F. Roca and C. Minarini (ENEA) for advice and support during the course of the activities. T. Varema and M. Kauk-Kuusik (TUT) are acknowledged for support in CZTS solar cell preparation.

REFERENCES

[1] X. Lin, et al., Energy Environ. Sci. 9 (2016) 2037.
[2] E. Lotter, et al., Proc. EU-PVSEC Munich (2016) 1156.
[3] F. Ringleb, et al., APL 108 (2016) 111904.
[4] S. Coppola, et al., APL 106 (2015) 261603.
[5] E. Mellikov et al., in: Copper Zinc Tin Sulfide-Based Thin Film Solar Cells, Wiley-VCH (2015) 289.