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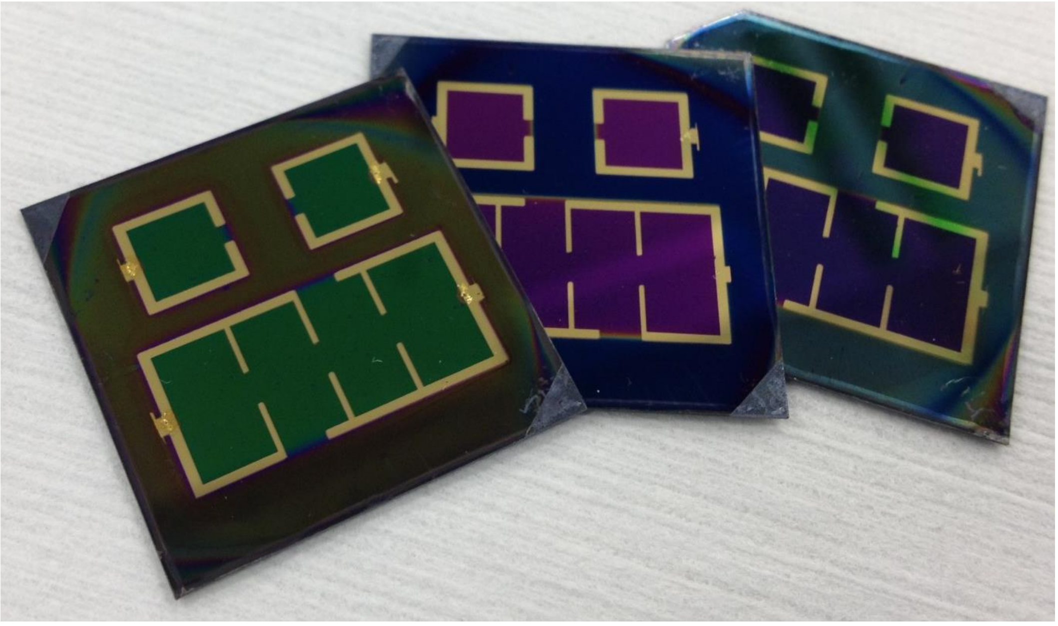
Final report

Systems for ultra-high performance photovoltaic energy harvesting (Synergy)



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List of abbreviations

CAPEX	Capital expenditures
CIGS	Copper indium gallium diselenide
CSEM	Centre Suisse d'Electronique et de Microtechnique
c-Si	crystalline silicon
Empa	Eidgenössische Materialprüfungs- und Forschungsanstalt
EPFL	Ecole Polytechnique Fédérale de Lausanne
EQE	External quantum efficiency
FA	Formamidinium
FF	Fill factor
J	Current density
J_{sc}	Short-circuit current density
LPI	Laboratory for Photonics and Interfaces
MA	Methylammonium
MPP	Maximum power point
PV	Photovoltaic
PV-Lab	Photovoltaics and Thin Film Electronics Laboratory
TFPV	Laboratory for Thin Films and Photovoltaics
V	Voltage
V_{oc}	Open-circuit voltage

Zusammenfassung

Viele etablierte Solarzellen-Technologien, darunter solche mit Absorber-Schichten aus kristallinem Silizium (Si) und Kupfer-Indium-Gallium-Diselenid (CIGS), erreichen Wirkungsgrade nahe ihrer Effizienzgrenze. Innovative Ansätze werden deshalb benötigt, um ihren Wirkungsgrad weiter erheblich zu steigern. Einer der vielversprechendsten Ansätze besteht darin, verschiedene Photovoltaik-Technologien zu Tandem-Solarzellen zu kombinieren. Dieses Projekt hat gezeigt, dass die noch junge Perowskit-Technologie mit bereits etablierten Technologien zu hoch-effizienten Tandemzellen kombiniert werden kann. Dafür wurden Perowskit-Zellen mit hoher Infrarot-Transparenz entwickelt und entweder mit einer Si-Zelle oder einer CIGS-Zelle kombiniert. Für aufeinandergeschichtete Tandemzellen wurden so Wirkungsgrade von 25.6 % (Si) und 22.7% (CIGS) erreicht. Zusätzlich wurden Perowskit-Zellen direkt auf Si-Zellen abgeschieden, um monolithische Tandemzellen herzustellen. Diese erreichten Wirkungsgrade bis zu 22.7% dank einer Ladungsträger-Rekombinationsschicht aus nano-kristallinem Si, welche die optischen Verluste minimierte. Die etablierten Photovoltaik-Technologien wurden für ihre Verwendung in Tandemzellen angepasst, indem ihre Infrarot-Spektralantwort verbessert wurde. Für CIGS-Zellen geschah dies vor allem durch Anpassen der Absorber-Schicht, bei Si-Zellen indem Elektroden mit hoher Transparenz in diesem Wellenlängenbereich entwickelt wurden. Die Skalierbarkeit der noch jungen Perowskit-Technologie wurde anhand von Modulen mit einem Wirkungsgrad von 16% für eine Fläche von 14 cm^2 gezeigt. Gegen Ende dieses Projekts wurde die Lebensdauer der Perowskit-Zellen erhöht, indem Verkapselungsverfahren und Perowskit-Materialien mit verbesserter Stabilität entwickelt wurden. Dieses Projekt war Teil eines gleichnamigen Nano-tera.ch Projekts mit Projekt-Nummer 530695, welches zusätzlich die Entwicklung von neuartigen Solarzellen mit Nanodrähten aus Galliumarsenid beinhaltete.

Résumé

Les technologies actuelles de cellules photovoltaïques, telles que celles à base de silicium (Si) ou de cuivre, indium, gallium et diselenide (CIGS), approchent leur limite d'efficacité. Des approches innovantes sont ainsi nécessaires pour dépasser cette limite. Ce projet démontre que de nouvelles technologies peuvent être combinées avec celles établies pour former des cellules tandems à haute efficacité. Dans cette optique, des cellules à base d'une perovskite, un matériau transparent dans l'infrarouge, ont été développées. Celles-ci ont été empilées mécaniquement sur des cellules de Si ou CIGS afin d'atteindre des efficacités de 25.6% et 22.7%, respectivement. Ces cellules pérovskites ont aussi été déposées directement sur la face avant de cellules Si afin de former des tandems monolithiques, démontrant des efficacités de 22.7%. Ces tandems monolithiques utilisent une jonction de recombinaison à base de silicium nanocristallin, ce qui permet de réduire les pertes optiques. L'optimisation des cellules photovoltaïques placées à l'arrière d'une tandem s'est concentrée sur l'amélioration de leur réponse spectrale dans le proche infrarouge. Pour les cellules CIGS, ceci a été atteint en modifiant la composition du matériau absorbeur, tandis que les contacts ont été rendu plus transparents pour les cellules Si. La fabrication de modules gravés par laser d'une efficacité de 16% pour une dimension de 14 cm^2 a démontré qu'il était possible de transposer à plus grande échelle la technologie des cellules perovskite. De plus, une méthode d'encapsulation robuste a été mise au point. La phase finale du projet s'est concentrée sur l'augmentation de la stabilité des cellules pérovskites, ce qui a été rendu possible en modifiant la composition de la perovskite. Ce projet a fait partie d'un projet Nano-tera.ch du même nom (numéro 530695). Celui-ci s'est aussi penché sur d'autres technologies solaires telles que celles à base de nanofils d'arséniure de gallium.

Summary

Established photovoltaic (PV) technologies, such as crystalline silicon (c-Si) and copper indium gallium diselenide (CIGS) solar cells, are approaching their practical efficiency limit. For further substantial performance improvements, disruptive approaches are therefore required. A highly promising approach lies in combining PV technologies to form tandem solar cells. This project demonstrated that the emerging perovskite PV technology can be combined with established technologies to form highly efficient tandem devices. Specifically, perovskite solar cells with high near-infrared transparency were developed and mechanically stacked onto either c-Si or CIGS cells. This led to tandem cell efficiencies of up to 25.6% and 22.7%, respectively. In addition, perovskite cells were directly deposited onto c-Si bottom cells, resulting in monolithic tandem cells with efficiencies of up to 22.7%. These monolithic tandems were made using a nano-crystalline Si recombination junction, which reduced optical losses. The optimization of the photovoltaic technologies used as bottom cells focused on the improvement of their near-infrared spectral response. For CIGS cells, this was reached by tuning the absorber layer composition, for c-Si cells by using highly transparent contacts. Up-scaling of the perovskite technology was demonstrated by fully-laser-scribed modules with efficiencies of up to 16% for an active area of 14 cm^2. In addition, a reliable encapsulation process for perovskite cells was developed. In the final phase of the project, major effort was dedicated to improve the stability of perovskite cells. By developing advanced perovskite materials, cells with strongly improved stability were obtained. This project was part of a Nano-tera.ch project with the same name (project number: 530695), which additionally included research on more prospective solar cell technologies, such as gallium arsenide nanowire cells.

1. Project goals

Since decades, the photovoltaic (PV) market has been dominated by wafer-based crystalline silicon (c-Si) solar cells, whose efficiency approaches the practical limit. In addition, thin-film solar cells based on copper indium gallium selenide (CIGS) compounds have recently reached efficiencies beyond 20% and are thus nearly as efficient as c-Si cells. As a result, further substantial improvements of these established industrial PV technologies require disruptive approaches. One of the most promising approaches for further performance improvement is to combine these single-junction cells with a high-bandgap PV technology to form tandem devices. In a tandem cell, the high-bandgap top cell absorbs the visible light from the solar spectrum, while transmitting the infrared light to the wide bandgap c-Si or CIGS bottom cell. This allows for a strong reduction of thermalization losses, enabling efficiencies well beyond those achievable by single-junction devices. Until recently, this concept has only successfully been employed for either very expensive or low-efficiency PV technologies. With the emergence of the perovskite PV technology, a suitable high-bandgap top cell candidate became available for highly efficient tandem cells when combined with c-Si or CIGS cells. Perovskite materials are particularly interesting for tandem applications, as they offer a sufficiently wide and tuneable bandgap, exhibit excellent electrical properties as well as low sub-bandgap absorption, and promise low fabrication costs. The *Synergy* project aims to combine perovskite top cells with c-Si or CIGS bottom cells to develop tandem solar cells with efficiencies beyond those achievable with state-of-the-art industrial PV technologies. This could enable ultra-high performance PV energy harvesting systems with the potential to provide both the terawatts of sustainable energy needed for future generations as well as ubiquitous wearable and electronic applications with integrated power source.

2. Work undertaken and findings obtained

The work presented below is part of the *Synergy* co-funded by the Swiss Office of Energy (contract number: SI/501072-01) and the Nano-tera.ch (contract number: 530695) programme. Several project partners (EPFL PV-Lab, EPFL LPI and Empa TFPV) are also part of the consortium of the thematically similar *NFP 70 Energiewende* project *PV2050: Building blocks for Next Generation Multi-Junction Solar Cells* (contract number 153916). Several results shown here were co-developed for both projects.

Perovskite solar cell with transparent rear electrode

For monolithic tandem integration with CIGS bottom cells, a perovskite top cell in the p-i-n configuration is required to enable recombination of holes with electrons generated in the CIGS bottom cell. Figure 1a shows a cross-sectional scanning electron microscopy image of a planar p-i-n near-infrared-transparent perovskite solar cell in substrate configuration, where the cell is illuminated from the film side. The p-i-n substrate configuration allows for direct growth of the perovskite top cell onto CIGS bottom cells to realize monolithic tandem solar cells. As shown in Figure 1b, the champion cell shows a steady-state efficiency of 16.8%, which is the highest value reported so far for substrate configuration perovskite solar cells. Additionally, the cell shows a high transmittance in the near-infrared region, making it suitable for tandem applications.

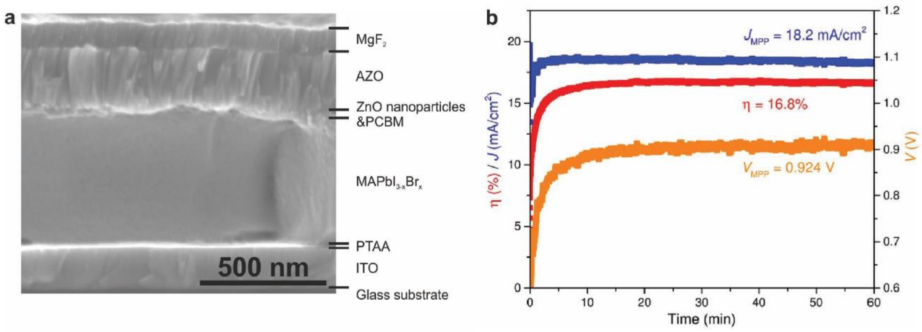

Figure 1: Cross-sectional scanning electron micrograph (a) and MPP measurement (b) of a p-i-n perovskite solar cell. Figure published in [1].

Besides power conversion efficiency, the long-term (>1000 hours) operational stability is an essential factor determining the economic viability of the near-infrared-transparent perovskite single-junction and perovskite-based tandem solar cells. A partial ion-exchange approach was developed at Empa to incorporate methylammonium bromide (MABr) into formamidinium (FA) lead iodide-based devices, yielding superior operational stability under continuous light soaking at elevated temperature. Figure 2 presents the stability of $FAMAPbI_{3-x}Br_x$ based solar cells. The non-encapsulated cell was kept at the maximum power point (MPP) condition under continuous one-sun illumination in N_2 atmosphere. The cell retained over 93% of its initial efficiency after 1000 hours light soaking at 60 °C as shown in Figure 2a. The current-voltage (Figure 2b) and external quantum efficiency (Figure 2c) characterization conducted at standard test conditions confirmed the superior stability and suggested that the degradation was primarily due to the loss of active area.

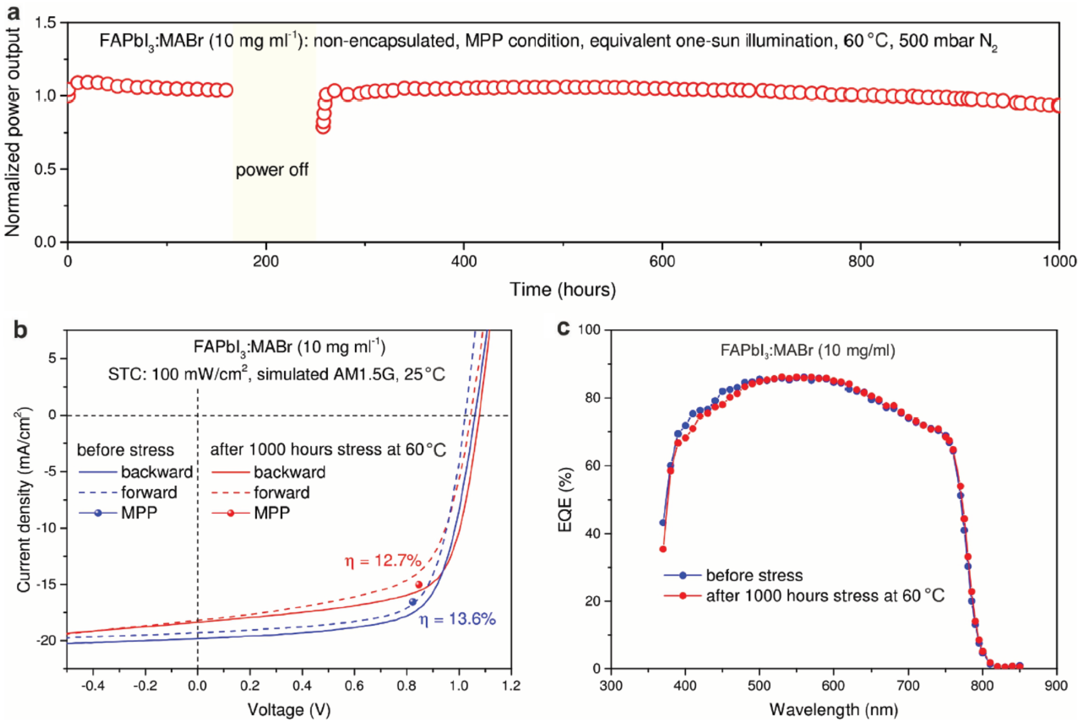

Figure 2: Long-term operational stability of $FAMAPbI_{3-x}Br_x$ perovskite solar cells. (a) Normalized efficiency measured under continuous 1-sun illumination at 60°C using a 500 mbar N_2 atmosphere. The current-voltage curves (b) and external quantum efficiency spectra (c) measured under standard test condition (25°C, AM1.5G, 1000 W/cm^2). Figure published in [1].

Fabrication and characterization of perovskite / c-Si tandem solar cell

Monolithic perovskite / silicon heterojunction tandem cells typically use a transparent conductive oxide as an intermediate recombination layer between the silicon bottom cell and the perovskite top cell. For tandem cells with a flat front side, this has the disadvantage of pronounced reflection losses due to a refractive index mismatch. CSEM in collaboration with EPFL PV-Lab developed a recombination junction based on a p-type / n-type nanocrystalline hydrogenated silicon layer stack. This layer stack has a refractive index, which is similar to that of the crystalline silicon wafer of the bottom cell, reducing reflection losses. After optimization of this recombination junction, tandem cells with an efficiency of up to 22.7% (current-voltage measurement) were obtained. The MPP tracking yielded a steady-state efficiency of 22.0%. This efficiency was higher than that of tandem cells with a transparent metal oxide recombination layer, resulting from an increase in short-circuit current of 1 mA/cm^2.

A major limitation of current state-of-the-art perovskite materials for tandem applications is their relatively low bandgap. Ideally, this bandgap should be ~1.7 eV to allow for matched photocurrent generation in the perovskite top cell and the silicon bottom cell. This is especially important for monolithic tandem cells, where the sub cell with the smaller short-circuit current is limiting the tandem cell current. However, also for 4-terminal tandem cells, increasing the top cell bandgap towards the ideal value can improve tandem performance by increasing the light transmitted to the bottom cell. EPFL PV-Lab developed a perovskite material with an increased bandgap of 1.65 eV, adding formamidinium and bromide to the commonly used methylammonium lead iodide material. With this modified perovskite material, 4-terminal tandem cell measurements were performed and efficiencies of 25.6% were obtained with a 0.25 cm² top cell and a 4 cm² bottom cell (Figure 3). This improvement compared to the previously reported 25.2% with a methylammonium lead iodide perovskite top cell was mainly due to an increased current in the bottom cell.

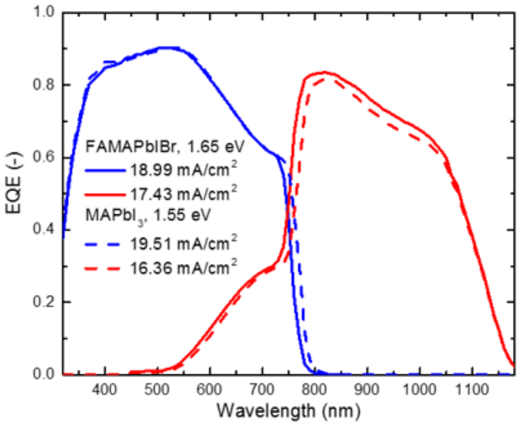

Figure 3: External quantum efficiency spectra of 4-terminal perovskite / silicon heterojunction tandem cell measurements. The top cell either contained a methylammonium lead iodide ($MAPbI_3$) or formamidinium methylammonium lead iodide bromide ($FAMAPbI_{3-x}Br_x$) absorber layer.

Fabrication and characterization of perovskite/CIGS tandem solar cells

Using the highly efficient p-i-n perovskite top cell presented above, a 4-terminal perovskite / CIGS tandem cell with an efficiency of 22.7% was developed (Figure 4). Main routes for further performance improvements of 4-terminal perovskite/CIGS tandem cells include bandgap engineering of the perovskite top cell and CIGS bottom cell, and improved optical coupling between the sub cells.

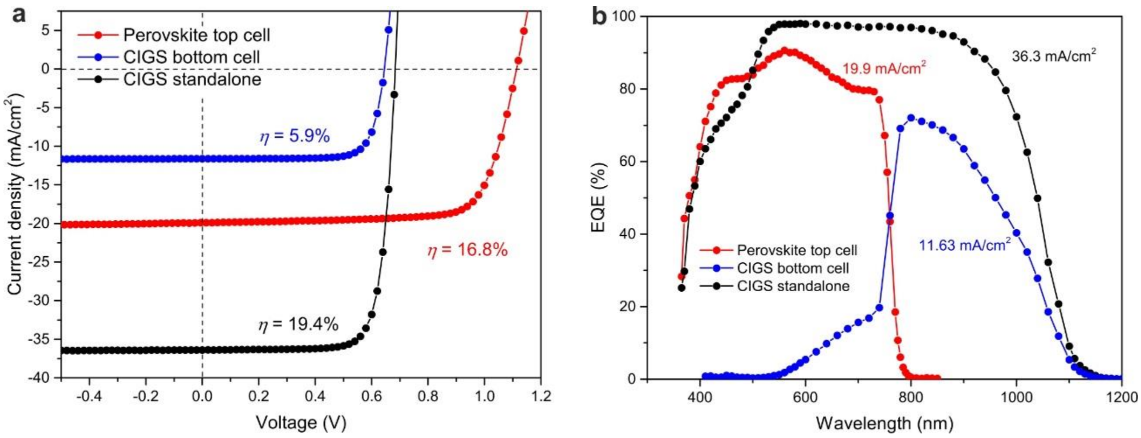

Figure 4: J-V curves (a) and EQE spectra (b) of 4-terminal perovskite / CIGS tandem cell. Figure published in [1].

In addition, EPFL LPI and Empa investigated monolithic multijunction devices composed of two series-connected CIGS cell connected in parallel with a near-infrared-transparent perovskite solar cell [2]. EPFL LPI developed perovskite absorber layers with bandgaps below 1.56 eV. When such a perovskite cell with an open-circuit voltage (V_{oc}) of 1180 mV is combined with two CIGS cells in this configuration, an efficiency of ~28% is reached for a wide range of V_{oc} values of the CIGS cells. In the event that the V_{oc} of the perovskite cell is further increased, efficiencies above 29% would be reachable.

Up-scaling and mini-module fabrication of perovskite cells

During the first years of the project, CSEM developed perovskite mini-modules based on a methylammonium lead iodide ($MAPbI_3$) perovskite absorber material. During the course of 2017, it became clear that this perovskite material had insufficient thermal stability. This is not only a major concern for the operational lifetime of the modules, but also does not allow for reliable encapsulation of the modules with typical industrial encapsulation schemes, which use temperatures of ~150°C.

Therefore, CSEM implemented a modified perovskite material to reach better moisture and temperature stability. Specifically, Cs+ and formamidinium (FA) ions were added and the iodide was partially replaced by bromide, leading to $CsFAMAPbI_{3-x}Br_x$ perovskite layers. With these more advanced perovskite materials, 14 cm^2 mini-modules with steady-state efficiencies of up to 16% were made (Figure 5).

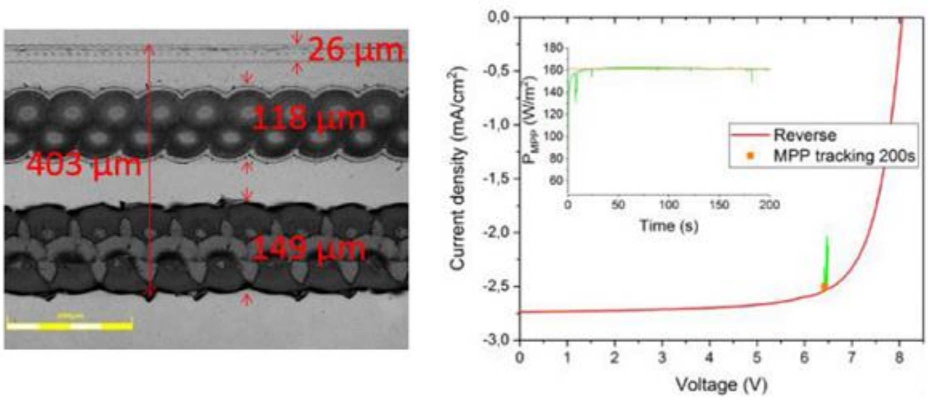

Figure 5: Microscopy image of the laser-scribed lines interconnecting the module segments (Left) and current-voltage curve of a seven-segment perovskite mini-module (right). The inset shows the output of that module during maximum power-point tracking. Figure published in [3].

CSEM in collaboration with EPFL PV-Lab also made substantial progress in up scaling monolithic perovskite / silicon heterojunction tandem cells. This was enabled by the nanocrystalline silicon recombination junction presented above, which by its low lateral conductivity strongly improved the cell's shunt resistance as compared to the previously utilized transparent conductive oxide recombination layer. This led to an efficiency of 18% for a monolithic tandem with an aperture area of 13 cm^2 (Figure 6).

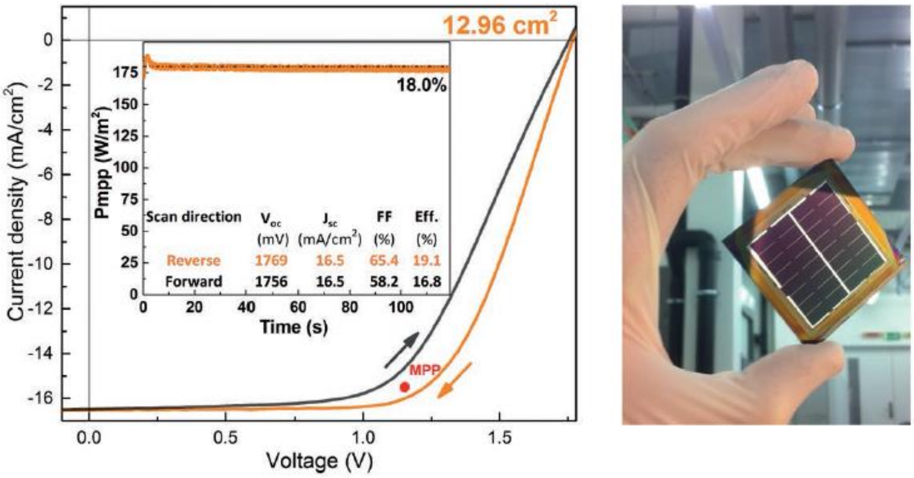

Figure 6: Current density-voltage curve, power output during maximum power-point tracking (left) and photograph (right) of a monolithic perovskite / silicon heterojunction tandem cell with nanocyrstalline silicon recombination junction and an aperture area of 13 cm^2. Figure published in [4].

Integration into industrial manufacturing processes

As production costs are a main concern for the industrial implementation of any new PV technology, a preliminary cost calculation was performed for perovskite / silicon tandem cells in the monolithic configuration [5]. As shown in Table 1, when assuming a performance gain of 4-5% as compared to the silicon single-junction cell and 4 additional layers deposited onto the bottom cell, tandem cells could become cost-competitive if no expensive materials are used in the perovskite top cell (e.g. organic charge transport materials or gold), costly modifications of the bottom cell are avoided, and the life time of the tandem cells remains similar to that of the silicon bottom cell. These calculations only take into account the additional steps induced by the top cell processing, with equipment that is depreciated over seven years. Consumable costs of 1.5 Euro/m^2 per step and 15% operational costs per year as fraction of CAPEX are assumed. For the area-related cost savings, a 18% nominal silicon single-junction module efficiency is considered.

Number of extra-steps	Invest-ment		Depre-ciation	Consum-ables	Opera-tional costs	Total process costs	Absolute efficiency gain	Manufactu-ring costs	Area related cost saving parks	Area related cost saving roof
	M€/step		€/m^2	€/m^2	€/m^2	€/m^2	%	€ct/W	€ct/Wp	€ct/Wp
3	1.5		1.61	4.5	1.69	7.79	4	19.5	2.7	6.4
4	1.5		2.14	6	2.25	10.39	4	26.0	2.7	6.4
3	3		3.21	4.5	3.38	11.09	4	27.7	2.7	6.4
4	3		4.29	6	4.5	14.79	4	37.0	2.7	6.4
4	2		2.86	6	3	11.86	5	23.7	3.3	7.6

Table 1: Results overview of preliminary cost calculation for monolithic perovskite/silicon tandem cells.

3. Conclusions

During the final stage of the project, significant progress was made. While the development of the CIGS and silicon heterojunction bottom cells ended in 2016, work was still ongoing to improve the perovskite top cell. By switching from the n-i-p polarity to the p-i-n one and by using multi-cation mixed-halide perovskite materials, considerable stability and performance improvements were reached, leading to near-infrared-transparent perovskite cells with an efficiency of 16.8%. Work on perovskite up-scaling led to mini-modules with efficiencies of up to 16%. Further improvements of perovskite / silicon heterojunction tandem cells led to 22.7% efficient monolithic tandem cells and 25.6% 4-terminal tandem measurements. Moreover, the development of a nanocrystalline silicon intermediate recombination layer enabled the upscaling of monolithic perovskite / silicon heterojunction tandem cells. Progress was also made on the development of 4-terminal perovskite / CIGS tandem cells, reaching efficiencies of up to 22.7%. The efficiency of this tandem cell configuration could be further enhanced by increasing the top cell bandgap and by improving the optical coupling between both sub cells. Finally, a preliminary cost calculation was made for monolithic perovskite / silicon tandem cells, indicating that this technology could become cost competitive.

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