

Options for Upscaling of Perovskite-Silicon Tandem Solar Cells

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Abstract

The efficiency of crystalline silicon solar cells is reaching its practical limit in R&D as well as in high-volume manufacturing. To further drive down the cost of electricity produced by photovoltaics, increasing the efficiency beyond the limit for single-junction crystalline silicon is strongly desirable. 2-terminal perovskite-silicon tandem solar cells represent an appealing option: they benefit from the installed manufacturing capacity and process maturity of the silicon bottom cell, and achieve the high efficiency potential by adding a thin-film top cell. The first lab results with efficiencies of more than 30% have recently been announced [1]. Naturally, such champion devices are initially produced on small areas of the order of 1 cm² and to date not all of the fabrication methods used in the laboratory meet the challenge of scaling up to industrial production. In this paper we discuss recent advances in the field, with a focus on potential device designs and manufacturing processes.

Introduction

The PV industry is expanding to ever-larger manufacturing capacities and crystalline silicon has taken the largest market share, marginalising competing thin-film technologies during this expansion. The ability to deliver a high power conversion efficiency has been a crucial factor leading to this success story. A high power

conversion efficiency reduces the overall levelised cost of electricity, which – together with the low degradation rates – are the central metrics with respect to investment decisions. With the achievable efficiency of crystalline silicon solar cells approaching the practical limit, further improvements can be achieved by increasing the number of light-harvesting absorbers. In this way, different sub cells, whose band gaps are tailored to the incident solar spectrum, can selectively harvest the photons and more efficiently transform the photon energy into electricity. Such multijunction cells have successfully been developed in R&D and are in commercial production for space applications and concentrator technology. While crystalline silicon has a fixed optical band gap, thin-film technology offers the ability to tune the band gap for the desired application. Therefore, the combination of crystalline silicon and thin-film technology could be the technology of the future.

One of the upcoming tasks is to select the best thin-film technology for combination with crystalline silicon. In recent years, the perovskite material class has attracted significant attention due to, first, the ease with which the band gap can be tuned by adjusting the composition and, second, a rather high defect tolerance. Also, it is potentially a low-cost technology because metal halide perovskites can be deposited from abundant materials using low-temperature processes and therefore it is at the centre of development efforts in academia and industry. In its simplest implementation, the multijunction concept uses two junctions and the result is a so-called tandem cell. This already offers a major increase in the theoretically achievable efficiency considering the radiative limit, which is boosted from 33.6% for a single-junction crystalline silicon solar cell to 45.2% for a tandem cell based on a crystalline silicon bottom cell (see Figure 1). By adding further junctions, the increase in efficiency becomes less pronounced. The practical efficiency limit of such multijunction concepts will also be governed by the losses arising from the need for optical and electrical coupling between the sub cells and the increasing process complexity and costs.

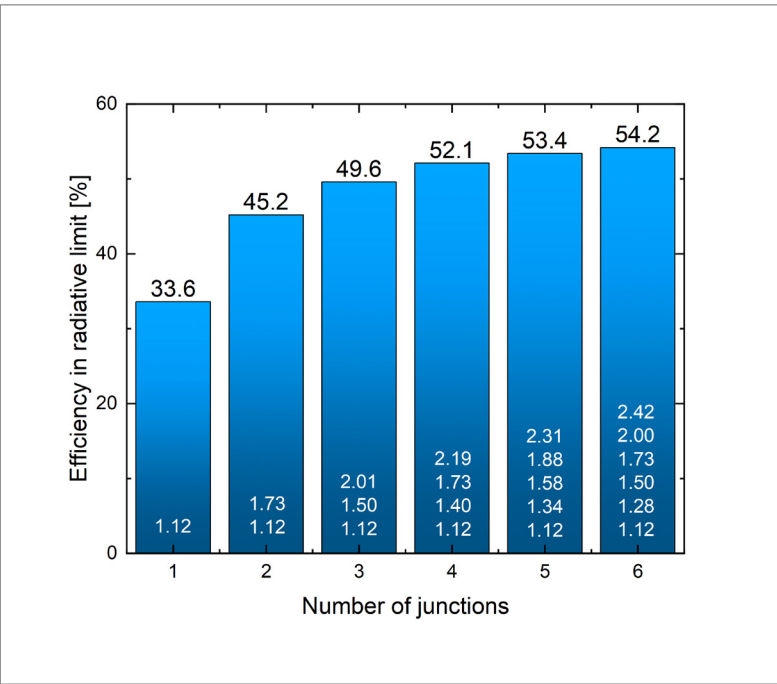


Figure 1: Obtainable efficiency in the radiative limit for crystalline silicon solar cells and multijunction cells with a crystalline silicon bottom cell. The white numbers inside the columns provide the optimum band gaps for the sub cells. Graph: provided by Patrick Schyglua; see also [2].

Also, a choice needs to be made for the electrical contacting scheme. In principle, every sub cell can have its own electrical contacts (one per polarity)

UPSCALING SOLAR EFFICIENCY

TACKLING GLOBAL CHALLENGES TODAY

VON ARDENNE 

2018
First R&D tool for
perovskite technology installed

2002
First production tool for c-Si PV

1996
First production tool for thin-film PV

GW STAGE

N-TYPE SOLAR TECHNOLOGIES
HETEROJUNCTION, TOPCON, IBC

R&D STAGE

PEROVSKITE TANDEM TECHNOLOGY

MAINSTREAM

PILOT STAGE

>43 GW
Provided capacity of
glass- & wafer-based PV

- ✓ Gigawatt production coating equipment for n-type solar technologies from the market leader
- ✓ R&D and pilot production equipment for Tandem PV
- ✓ Advanced equipment technology for PVD and evaporation at all scales of production from lab to fab with competitive costs per watt

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for connection in a PV module. For a tandem cell, this would be a 4-terminal device in which the sub cells are only optically coupled to each other. There is also a configuration where the two cells share a common contact, hence resulting in a 3-terminal device. Alternatively, a 2-terminal tandem is an option, where, similar to a silicon solar cell, the electrical energy is only extracted at two external contacts and where the current generated in the sub cells should be identical for optimum performance (see Figure 2).

While the latter condition – matching of the current in both sub cells – imposes some technological constraints, it is the least complex configuration when connecting several cells in a PV module. Hence, 2-terminal tandem cells with a crystalline silicon solar cell on the bottom are a natural starting point and are the focus of this article.

Device architecture of 2-terminal tandem solar cells

When integrating two sub cells with different band gaps into a single device, the cells need to be adjusted to each other because in a 2-terminal

device the cells are not only optically but also electrically coupled. In the following, we address the choice of the bottom cell and the individual layers of the top cell. For example, Figure 3 shows a schematic sketch of a perovskite-silicon tandem cell with an HJT bottom cell.

Bottom cell

There are several choices for the bottom cells, even when constraining ourselves to crystalline silicon as the absorber material, like PERC, TOPCon, HJT, IBC or its variants. For the best performance, the cell should be optimised to harvest the long wavelength photons transmitted by the top cell and feature very well-passivated surfaces. Parasitic absorption in the short wavelength of the solar spectrum is, however, less relevant, since those photons have already been absorbed in the top cell. With the photocurrent being about half of a single-junction silicon cell, the tandem cell is more tolerant to high series resistance, but low-light-intensity performance becomes even more relevant.

Finally, the front side should be textured, as it is with single-junction crystalline silicon. The light

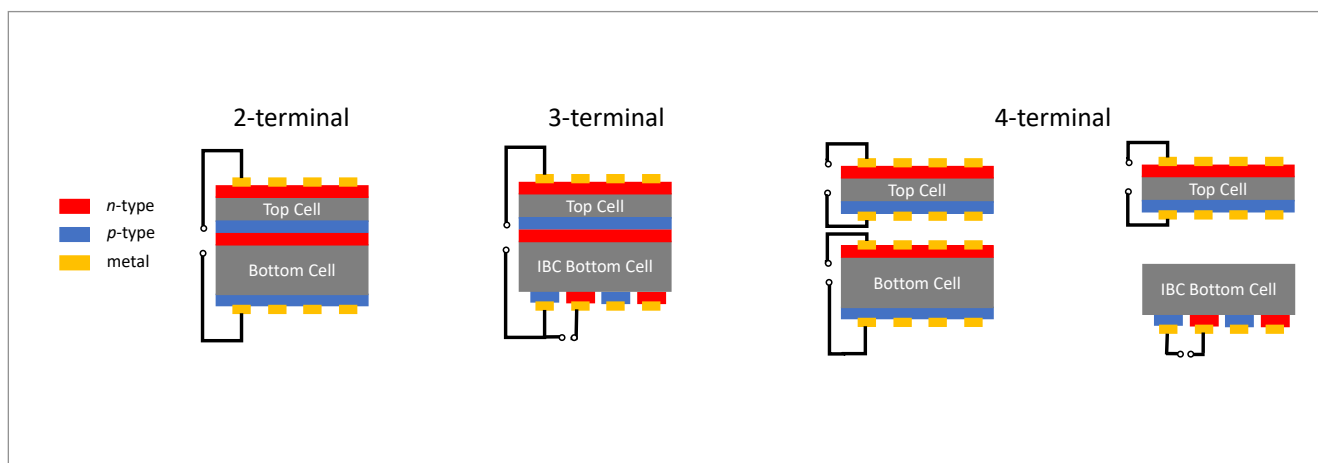


Figure 2: Sketch of tandem cells with two, three or four external contacts. Notably, a 2-terminal configuration requires current matching between both sub cells for optimum performance, while 3- and 4-terminal devices are only optically coupled yet more complex in terms of PV module assembly and maximum power point tracking.

is coupled in effectively and the photons have a longer path length within the tandem cell. A texture on the rear additionally supports the light trapping but is only mandatory when the front side is planarised. Also, a texture on the front side significantly improves the annual energy yield in fixed-tilt installations [3]. However, it is more challenging from a technological point of view to achieve conformal thin film deposition on a textured surface. This imposes technical constraints for the choice of deposition methods, as discussed in more detail later.

Regarding the choice of the best bottom cell, many factors need to be considered. From a commercial perspective, using a standard PERC cell as a bottom cell would likely be the fastest way to achieve large-scale adoption of the tandem concept. However, the integration of a very low-temperature processing route for a perovskite top cell into an overall attractive process flow might prove more complex than initially envisioned. This is because PERC cell manufacturing has been optimised for high-temperature metallisation and fire-through of metal pastes through electrically insulating layers, whereas for a 2-terminal tandem cell, the sub cells also need to be electrically connected to each other through conductive layers. Hence, a full-area passivating front contact of the silicon cell that enables vertical carrier flow, like TOPCon or POLO, can offer a performance benefit [4,5]. The surface passivation is superior to PERC cells yet conductive, which enables the interconnection of the sub cells without patterning process steps. For interconnection to the top cell also a polycrystalline tunnel junction can be employed [10]. While the highly doped poly-silicon layers are rather absorptive in the short wavelength spectrum, this does not compromise the efficiency in a tandem cell because the short wavelength photons are already absorbed in the top cell. However, the established technology for

this intermediate interconnection layer is still a recombination TCO.

IBC cells are also amongst potential device structures for perovskite-silicon tandem solar cells. Since an IBC cell has the contacts for the two polarities already on its rear side, a 3-terminal device can be built. Because of the additional third terminal, the requirement for current matching of the two sub cells is lifted. However, for a PV module, the interconnection of the cells in a full-size module is complex when exploiting the third terminal and the practical impact on energy yield depends on the actual circumstances [7].

While a TCO layer may also be applied onto the front side of an IBC, PERC or TOPCon cell [8], an HJT bottom cell naturally already ends with a TCO layer on the front. Moreover, process technologies for HJT and perovskite cells are often very similar (e.g. vacuum equipment). Hence, HJT bottom cells are especially prominent in 2-terminal perovskite-silicon tandem cell development. Also, an HJT cell's major weak point is often the relatively strong parasitic absorption of short wavelength photons – in a tandem cell, these high energy photons are already absorbed in the top cell, hence this disadvantage becomes unimportant. In combination with a rather simple process flow for a lab environment, these factors have led to HJT becoming the workhorse for 2-terminal perovskite-silicon tandem solar cells, at least in R&D.

When factoring in manufacturing costs and yield in high-volume manufacturing, this might change, and further development efforts will continue with many variants of silicon solar cell device structures as the bottom cell.

While all discussed bottom cell types are already in PV mass manufacturing, this is not yet the case for all layers of the top cell. The function and examples of the individual layers are discussed in more detail in the following.

Interconnection layer

In monolithic series connected tandem solar cells, the interconnection layer electrically connects the sub cells. Ideally, this is done without voltage losses at the transition between the sub cells and without parasitic absorption in the interconnection layer for infrared light, which should be absorbed in the silicon bottom cell.

Such an interlayer prevents the formation of a blocking pn-junction between the hole contact of the top cell and the electron contact of the silicon cell. Conventionally, either a recombination layer or a tunnel junction are used. Tunnel junctions are widely and very successfully used in III-V multijunction cells [9]. The tunnel junction itself is made by one degenerately doped n++ layer and one p++ layer, and the carrier transport takes place as tunnel current above the very narrow depletion layer. Such a tunnel diode can be realised, e.g., by PECVD deposited and annealed poly-Si layers, as shown by Luderer et al. [10].

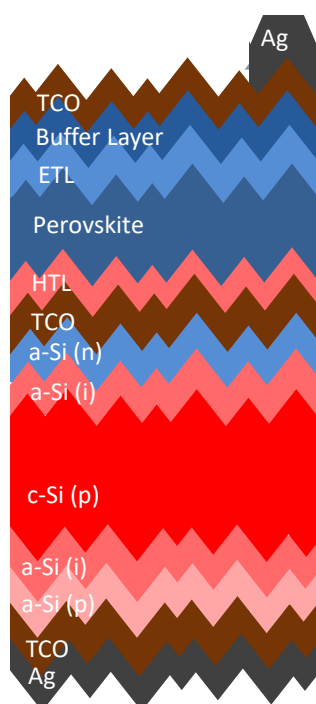
Alternatively, so-called recombination layers can be used. These layers act like a conductive layer, which can transport electrons and holes. So, the electrons from the silicon cell recombine with the holes from the top cell. The simplest example is evaporated ultra-thin metal films, which are used,

e.g., in organic-based tandem cells [11]. These metal layers have to be thin enough to avoid parasitic absorption. An often-used recombination layer in a Pero-Si Tandem solar cell is ITO or other metal oxide layers [12], which can be easily deposited by sputtering. ITO is a good conductor for electrons and holes, which is why it is used for both polarities in silicon heterojunction solar cells. The thickness of the metal oxide layer should also be thin enough, not only for low absorption, but also to avoid optical interference effects, leading to unwanted reflections.

Several different options for industrial depositions of the interconnection layer are available, and the choice largely depends on the overall device design and process integration.

Hole Transport Layer (HTL)

Depending on the deposition sequence, the top cell configuration is either a n-i-p or p-i-n structure. First reports on pero-si tandem devices were built in the n-i-p layer sequence but poor optics caused by a thick layer of the hole transport material Spiro-OMeTAD limited device performance. Since 2017 most publications focus on the inverse p-i-n structure, where the hole transport layer (p-type) is deposited first. In the following we focus on this particular structure, however, it is noteworthy that the choice of



Buffer Layer, e.g. SnO , Al_2O_3 , etc.

ETL: Electron Transport Layer C60

HTL: Hole Transport Layer, e.g. NiO , SAM, etc.

TCO: Transparent Conductive Oxide e.g. ITO, AZO, IZO, etc.

Figure 3: Sketch of a textured perovskite-silicon tandem solar cell with a p-type HJT bottom cell, as used for development at Fraunhofer ISE. There are several options for the choice of materials and their deposition methods, and these are being continuously investigated in the PV community.

p-i-n vs n-i-p depends on the development of suitable charge transport materials. Common examples for suitable hole transport layers in the p-i-n structure are wet chemically applied self-assembling monolayers (SAM) such as 2PACz and Me-4PACz [13,14].

Moreover, vacuum-based processes are suited for conformal coating on textured surfaces. Thin evaporated Spiro-TTB [15] has been reported, however, they show film degradation in the form of melting into the valleys (due to low glass transition temperature) upon temperature treatment above 150°C [17]. Such temperature is commonly used for thermal annealing of the perovskite layer on texture as well as curing of metal paste. As an alternative, sputtered NiOx can be deployed, most likely in combination with a subsequent deposition of SAMs [16] (for improved stability and minimized surface recombination).

Evaporation of organic compounds or sputtering of metal oxides are methods that can be adopted to the requirements of wafer-based high-volume manufacturing.

Perovskite

The deposition of the perovskite absorber on a large wafer area can be accomplished by several methods. Spin-coating is a very versatile technology, but it is restricted to small areas and only utilises a small fraction of the prepared chemical solution. Slot-die coating is not only used on perovskite single-junction thin-film modules but can also be used for silicon-based perovskite-silicon tandem solar cells [18]. However, when applied on textured silicon, the texture is basically filled up and the perovskite surface is mostly planarised [19].

Two methods for a conformal coating of textured silicon that preserve the texture are co-evaporation [20,21] and the hybrid route combining co-evaporation and wet chemical processing [22]. They have shown very promising results that are likely to be scalable to a large wafer area and also could be envisioned to be manufacturable on an industrial scale [23].

In the hybrid route, the deposition of the perovskite is split into two process steps.

In the first step, the inorganic components are thermally evaporated to form a scaffold, allowing the conformal coating of pyramids of arbitrary size and shape. In the second step, the conversion into perovskite is accomplished by infiltration with organic salt solutions. For the second step, several deposition techniques have been evaluated by researchers and are already used in industrial fields

like spray coating, inkjet printing and slot-die coating (see Figure 4).

The benefit of this approach is that a conformal coating of the pyramids can be achieved and the texture is preserved.

Thermal annealing at temperatures around 130–150°C completes the conversion process and the formation of perovskite. The difficulty with this hybrid process flow is to ensure a good infiltration of the organics into the scaffold and complete conversion into a high-quality perovskite crystal without remnant PbI₂ at the lower interface. Alternatively, the entire perovskite can be evaporated in a single process step [21,22] (co-evaporation of inorganic and organic components). Alternatively, it can even be achieved sequentially, where the inorganic layer is deposited first and an organic layer is subsequently deposited on top [24]. Such two-step processing minimises the use of expensive and potentially harmful solvents in the process flow and separates the evaporation of the organic materials from the inorganic ones, which may be a significant advantage from an equipment and process stability point of view.

Electron Transport Layer (ETL)

For efficient current collection, an ETL must be deposited. C60 can be evaporated and serves the purpose reasonably well. However, parasitic absorption is high and the interface is not well passivated, leading to significant losses in all solar cell parameters. To improve this interface, an additional layer can be introduced between the perovskite and the C60. LiF has been shown to improve the passivation by a field effect [25], but the stability of the cells is severely compromised. Recently, Liu et al. [26] proposed MgF₂ as an alternative, with good results and strongly improved stability. It appears that it is less important which layer exactly is deposited, so long as there is a thin layer between the perovskite and the C60 in order to prevent direct contact between the two, similar to the concept of contact displacement in the metal–silicon contact of high-efficiency silicon solar cells [27].

C60 is evaporated and this is, in principle, a scalable process for mass manufacturing. Despite its shortcomings with respect to absorption and recombination, it still appears to currently be the state-of-the-art material for the ETL.

Transparent Conductive Oxide (TCO)

Because of the extremely low conductivity of the bulk perovskite, lateral electrical transport is hampered and a conductive, yet transparent, layer must be placed on top. ITO is the most prominent candidate due to its track record in HJT cell technology. However, cost considerations and

limited availability present strong drivers towards the choice of alternatives like AZO or bi-layers [28]. Fortunately, developments from HJT technology aim in the same direction of improving transparency and conductivity. Furthermore, sputtering technology to deposit TCOs is already established for solar cell production in high-volume manufacturing.

Buffer layer

Sputtering of a TCO can result in significant sputtering damage to the ETL. To avoid or at least reduce the damage, a thin buffer layer may be

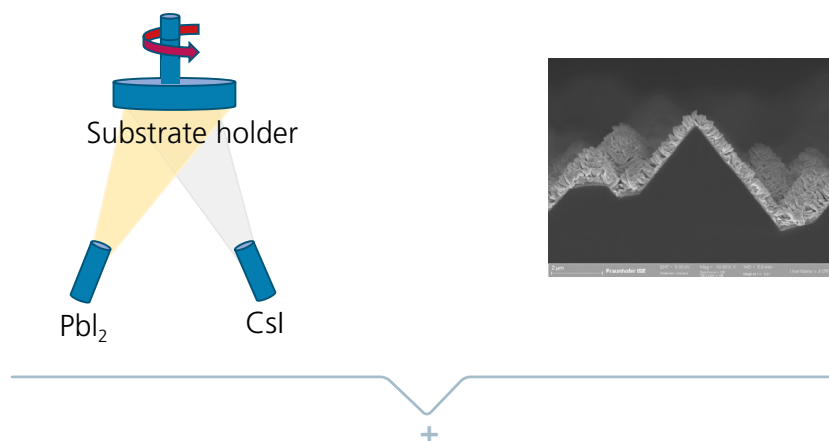
placed between the C60 and the TCO, like SnOx [29]. Such thin layers may be deposited by ALD, which has been used for the deposition of Al₂O₃ in industrial PERC processing.

Metallisation

In lab environments where the test cells are small (of the order of square centimetres), usually narrow traces of thin silver are evaporated as a front electrode. Alternatively, and suitable for an industrial process, screen-printing of ultra-low temperature silver paste can be employed. The

Step 1

Thermal evaporation of PbI₂ and CsI.



+

Step 2

Infiltration with organic salt solution via spray coating, inkjet printing or slot die coating (or spin coating for reference)

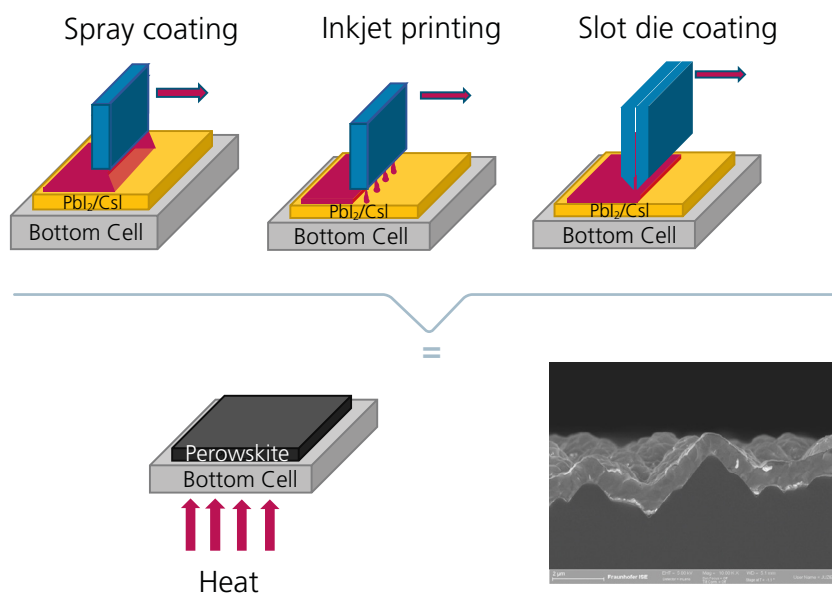


Figure 4: Hybrid process route for conformal coverage of a textured silicon bottom cell. Step 1: Deposition of a thin film of the inorganic components (e.g. CsI, PbI₂, CsBr or PbBr₂). Step 2: Exposure of the inorganic thin film to the organic component from solution. After subsequent thermal annealing the precursors are converted to the perovskite thin film. The thin film is converted to perovskite.

paste must adhere to the surface and provide good electrical contact and line resistance, even though the curing temperature is limited to about 150°C to prevent damage to the perovskite top cell with all its layers. This low-temperature requirement puts severe constraints on the paste composition and printability; it even exceeds the requirement of HJT cells. Screen-printing of silver paste is the standard method for the metallisation process in the PV industry. It has successfully been applied on perovskite-silicon tandem solar cells [30,31] and upscaling to larger areas has also successfully been demonstrated [23].

Anti-reflection coating

The standard process for the anti-reflection coating on single-junction crystalline silicon solar cells consists of a silicon nitride film with a refractive index of about $n_{\text{SiNx}} \sim 2$ by PECVD. This is in between the refractive indices of the encapsulant/glass of $n_{\text{glass}} \sim 1.5$ on the front side and the silicon wafer $n_{\text{silicon}} \sim 4$, and effectively

improves incoupling of sunlight. In a perovskite-silicon tandem cell, however, the refractive index of the perovskite is much lower than silicon $n_{\text{perovskite}} \sim 2.5$ and there is no need to add an ARC for perovskite-silicon tandem solar cells for use in encapsulated PV modules [32].

Whether a more efficient PV module results in a lower cost of electricity depends on, besides other factors, whether the additional efficiency is obtained in a cost-effective way. In the following cost calculations, we shed light onto the relationship of cost and efficiency increase based on certain assumptions.

Cost calculations

As mentioned before, for large-scale implementation of perovskite-silicon tandem solar cells, viable bottom cell candidates are PERC, TOPCon, SHJ and IBC. Wu et al. [4], e.g., published results for a device based on an industrial TOPCon bottom cell process route, achieving 27.6%.

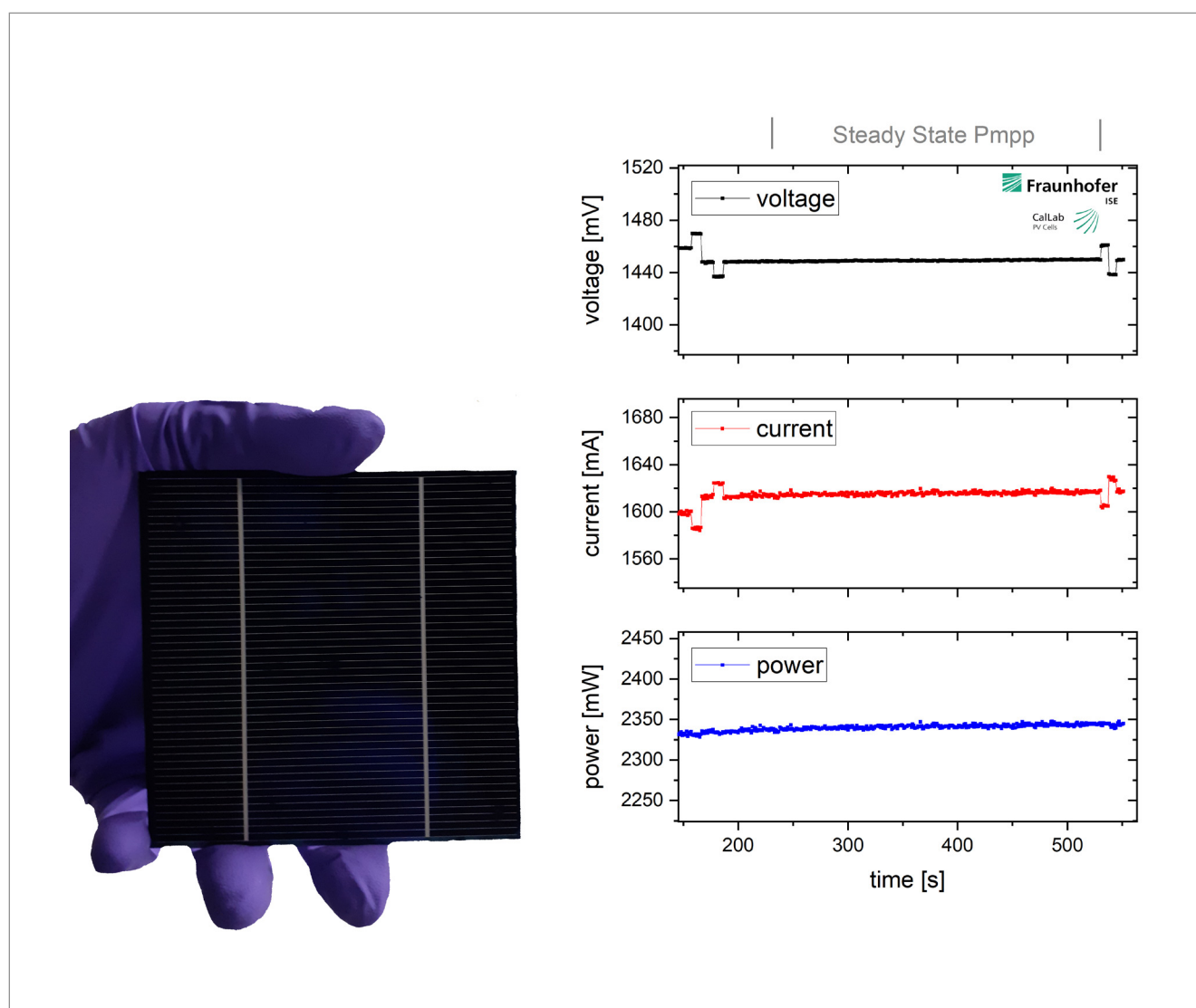


Figure 5: 104 cm² perovskite-silicon tandem solar cell with screen-printed front metallisation fabricated at Fraunhofer ISE. Power conversion efficiency of 22.5% under steady-state measurement conditions certified by Fraunhofer ISE Callab Cells.

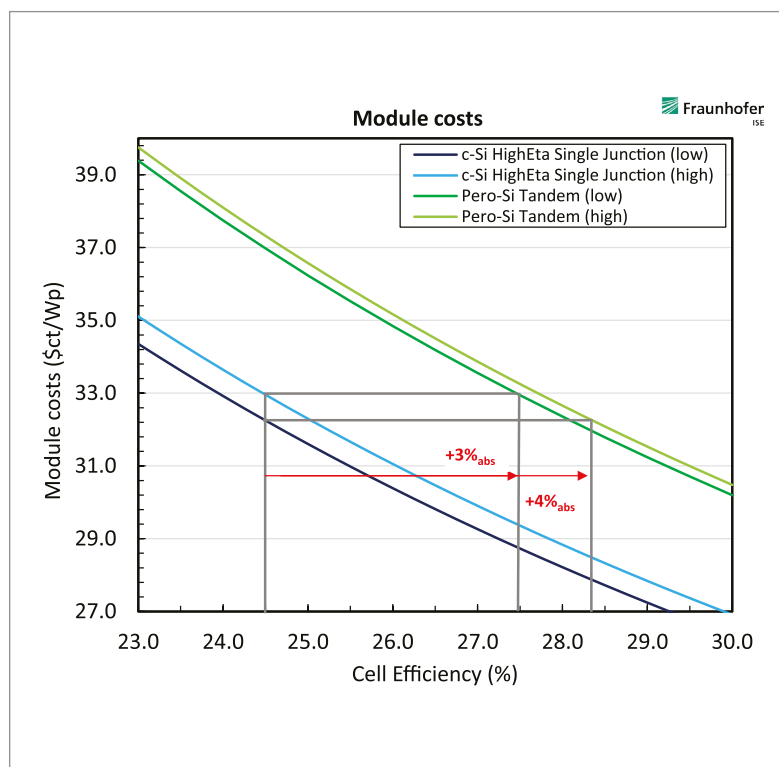


Figure 6: Impact of the cell device efficiency on the module cost for the total cost of ownership calculation. For a cell efficiency of 24.5% for single-junction devices, an efficiency advantage of 3–4%_{abs} is required for the tandem cells to reach the same module costs. The terms “low” and “high” refer to the low-cost or high-cost options, respectively, within the investigated device structures and process flows.

Messmer et al. [33] compared PERC, TOPCon and SHJ to potentially serve as silicon bottom cell devices, based on device simulations and a bottom-up total cost of ownership assessment. Updated cost calculations at Fraunhofer ISE, using M10 TOPCon or HJT as bottom cells, show that with a PVD-based hybrid route for the processing of the perovskite top cell, the required CAPEX investment for tandem cell production equipment increases by 40–90% over a single-junction counterpart for a 10 GWp factory. The operational costs (OPEX) for the tandem cell production (without depreciation and costs for the silicon wafer) increase by 50–90%. However, the high levels of uncertainty regarding economies of scale for the current low-volume perovskite top cell materials significantly impact the OPEX of the top cell device.

Assuming an M10 silicon n-type wafer price of 1 USD per piece currently (December 2022), a cell efficiency advantage of 3–4%_{abs} for the tandem cells would be required to reach a similar full cell cost per Wp to the standalone c-Si single-junction devices.

Integrating the tandem cells into a module requires additional encapsulation measures to be taken into account for assuring device stability. Assuming these measures correspond to additional module production costs (without the cells) of about 20%, with the same cell-to-module loss/

gain, the full module production costs for the tandems would reach the same level per Wp as the c-Si single-junction modules. Thus, our total cost of ownership calculations show that a module efficiency advantage of 3–4%_{abs} for the tandem devices leads to similar production costs per Wp for the module level as for the c-Si single-junction technologies (see Figure 6).

The tandem-related efficiency benefit transfers further to the costs downstream of the PV-system level, since with the same PV-system area and balance-of-system (BOS) related costs, more system capacity can be installed. In the end, the levelised cost of electricity (LCOE) will remain the final measure of the viability of a new cell technology.

Conclusion

Monolithic perovskite-silicon tandem solar cells offer an attractive option for further efficiency increases in photovoltaic modules. The device design and process flow are still under intense development. Several potential solutions have emerged and are promising candidates for successful product development on a commercial scale. While the production equipment for perovskite solar cells is different from traditional crystalline silicon cells, some processes like evaporation or sputtering can already be found in the thin-film PV segment or in HJT production lines. Hence, part of the upscaling effort might be accomplished by modification of existing tools. For a successful launch of perovskite-silicon tandem solar cells, the efficiency increase at moderate cost will need to be proven on a full wafer area and on production tools capable of high-volume manufacturing. The perovskite-silicon tandem cells will have to demonstrate bankability and stability in the field for successful commercialisation. With the progress achieved to date and the current investment for R&D from the public sector as well as from industry, it should only be a matter of time before PV modules make the next major efficiency leap forward.

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About the Authors



Oliver Schultz-Wittmann received his doctoral degree in 2005 from the University of Konstanz after completing his studies at Fraunhofer ISE which resulted in the first 20% efficient multicrystalline silicon solar cells. In 2009 Oliver co-founded the California start-up TetraSun which developed polysilicon passivating contacts on thin oxides and copper plating for crystalline silicon solar cells. TetraSun was bought by First Solar and built a 100MWp line in Malaysia in 2015. After spending two years with a global project developer Oliver returned to Fraunhofer ISE in 2021 where he is currently leading a project for the upscaling of perovskite-silicon tandem solar cells.



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Martin Hermle received the Diploma degree in physics from the University of Karlsruhe, Germany, in 2003 and the Ph.D. degree in physics from the University of Konstanz, Germany, in 2008. He joined the Fraunhofer-Institute for Solar Energy Systems, Freiburg, Germany, in 2002. Since 2008, he has been the Head of the Department of advanced Development of High-Efficiency Silicon Solar Cells. His research interests include the development of solar cell technologies for high-efficiency silicon and tandem solar cells and the analysis, characterization and modelling of them. He has made important contributions to the development and understanding of passivating contacts (e.g. TOPCon) and tunnel diodes. Important achievements of his department are the first 26 % both side contacted silicon solar cells as well as their contribution to the highest efficient 35.9 % 2-terminal tandem solar cells with a silicon bottom solar cell.



Patricia S. C. Schulze is scientist and vice group leader of the group Novel Solar Cell Concepts at Fraunhofer ISE. She studied Chemistry at the University of Mainz, Leipzig, and Complutense de Madrid. 2020 she received the Ph.D. from University of Freiburg. Since 2016 she is exploring perovskite absorbers and contact materials for the use in monolithic perovskite silicon tandem solar cells at Fraunhofer ISE. Her focus is on investigation of thin films, development of fabrication routes, as well as optimization of high-efficient tandem and multi-junction perovskite-silicon devices.



Juliane Borchert is the head of the research group "Novel Solar Cell Concepts" at the Fraunhofer Institute for Solar Energy Systems as well as the head of the junior research group "Optoelectronic Thin Film Materials" at the University of Freiburg, Germany. She studied physics in Berlin, Groningen, and Halle (Saale). Her PhD research was conducted at the University of Oxford where she focused on co-evaporated perovskites for solar cells. She continued this research as a postdoctoral researcher at the University of Cambridge and AMOLF research institute in Amsterdam. Now she leads a team who are on a mission to develop the next generation of solar cells combining novel metal-halide perovskite semiconductors and established silicon technology into highly efficient tandem solar cells.

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